SESSION 5

WASTE MANAGEMENT

Tuesday: August 25, 1992
Co-Chairmen: D. W. Moeller
W. R. A. Goossens

OPENING COMMENTS OF SESSION CHAIRMAN MOELLER

EXPERIMENTAL STUDY ON THE VOLATILE RUTHENIUM DECONTAMINATION FACTOR OF THE PERFORATED PLATE COLUMN SCRUBBER
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MEASUREMENT OF CESIUM EMISSIONS DURING THE VITRIFICATION OF SIMULATED HIGH LEVEL RADIOACTIVE WASTE
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CLOSING COMMENTS OF SESSION CO-CHAIRMAN MOELLER

OPENING COMMENTS OF SESSION CO-CHAIRMAN MOELLER

On behalf of the Program Committee and the co-chair, Dr. W. R. A. Goosens, it is my pleasure to welcome you to Sessions 5 and 6 of the DOE/NRC Nuclear Air Cleaning and Treatment Conference. Session 5 is directed to "Waste Management," and Session 6 will be on "Instruments and Sampling." The papers presented during these two sessions will cover three subjects: (1) Radioactive waste management; (2) Instruments and sampling of air systems; and (3) Filter performance and testing.

Our first speaker for Session 5 is Dr. M. Kitamura of the Ishikawajima-Harima Heavy Industries, Japan. Dr. Kitamura will be speaking on an "Experimental Study on the Volatile Ruthenium Decontamination Factor of the Perforated Plate Column Scrubber."

NOTE: Dr. Moeller chaired Session 5 (covering papers 5-1, 5-2, and 5-4); Dr. W. R. A. Goosens chaired Session 6 (covering papers 6-1, 6-2, 6-3, 15-6 and 15-1).

EXPERIMENTAL STUDY ON THE VOLATILE RUTHENIUM DECONTAMINATION FACTOR OF THE PERFORATED PLATE COLUMN SCRUBBER.

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Abstract

During a high temperature treatment of the radioactive wastes that contain ruthenium, some ruthenium is volatilized and released to the off gas system. The volatilized ruthenium is removed with a scrubber and an adsorber. It was expected that the scrubber was one of the most effective equipments to remove ruthenium and the decontamination factor of the scrubber was studied.

The experimental apparatus was simulated a part of an actual perforated plate column scrubber. The non-radioactive ruthenium which simulated the radioactive ruthenium was volatilized and fed into the scrubber. The decontamination factor of the scrubber was determined by the ratio of the ruthenium concentration at the inlet and outlet of the scrubber off gas stream. The results showed that the scrubber removed the volatile ruthenium effectively as expected.

I. Introduction

There are some kinds of radioactive wastes generated from the nuclear fuel cycle facilities. They are defined as high level, medium level and low level waste depending on the activity. Some of them are liquid wastes and others are solid wastes. The radioactive ruthenium is sometimes contained in these wastes. In order to reduce the volume of the waste, high temperature processes are established. Among them, the incineration, evaporation and vitrification[1] may release the volatile ruthenium when they process the ruthenium contaminated waste. In such process an oxidizing and high temperature atmosphere tends to produce the volatile ruthenium.

Most of the off gas processes have the first scrubber as a quencher to recover the vaporized liquid and to cool the off gas, and then the volatilized ruthenium is also recovered simultaneously. However, after the treatment in the scrubber the small part of the volatile ruthenium may still exist in the off gas stream in case of conservative evaluation. The Silica-Gel adsorber is used for removing these remaining ruthenium, but it brings solid waste. In order to avoid generating the solid waste, the water scrubbing process was proposed and studied. The previous study showed that the water scrubbing was the one of the effective processes to remove the volatile ruthenium.[2] Therefore, the present study simulated a NOx absorbing scrubber in the vitrification off gas system.

This study evaluated the performance of the water scrubber to remove the ruthenium from the off gas stream and the stability of it through the series of studies with changing experimental conditions. In these studies the inlet and outlet ruthenium concentration in the off gas were measured and the decontamination factor(DF) value was calculated.

II Experimental Method

Experimental apparatus

The experimental apparatus consists of the scrubber and auxiliary equipments. The scrubber is a perforated plate column with three plates and it simulates a part of an industrial scaled plate column. The diameter of the column is 0.2m and height between plates is 0.4m that is almost the same as that of the industrial column. The column installs nozzles, a gas inlet and outlet, a scrubbing water inlet and outlet. Figure 1 shows the schematic of the scrubber.

The auxiliary equipments consist of a volatile ruthenium generator, a gas supplying system, a scrubbing water supplying system and ruthenium samplers. Figure 2 shows the schematic of the scrubber and auxiliary systems.

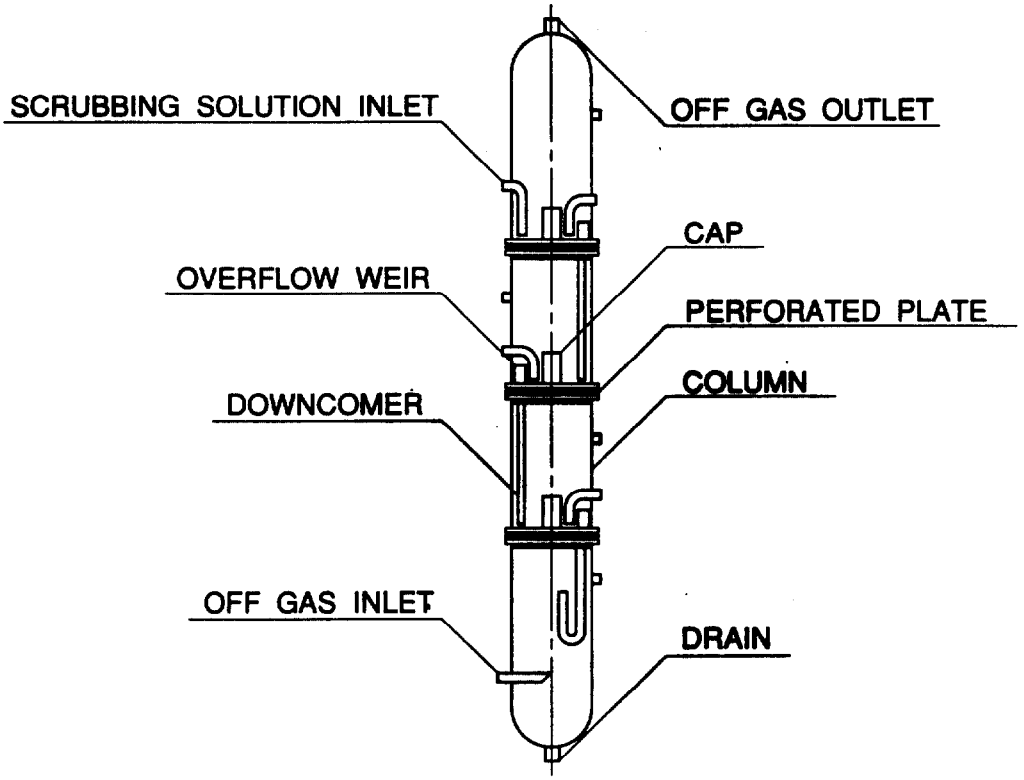
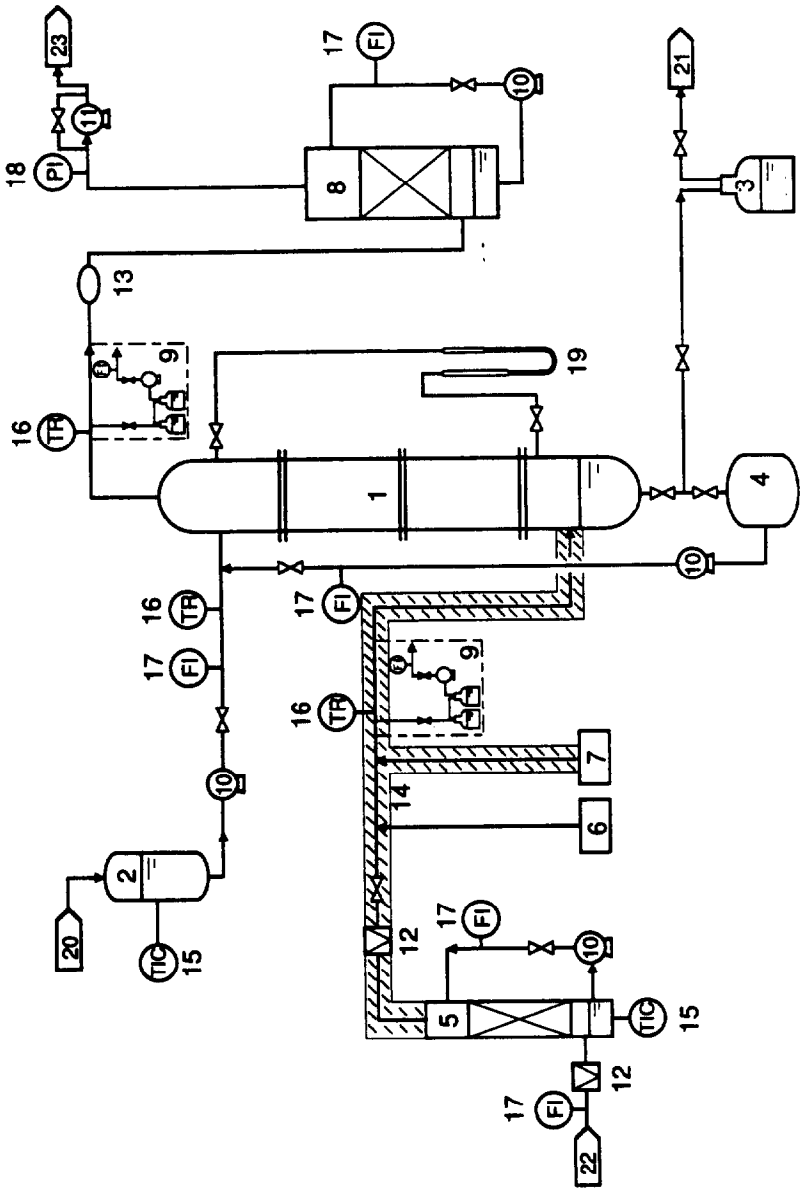


Figure 1 Schematic of the experimental scrubber.



(1) SCRUBBER
(2) FEED TANK
(3) DRAIN TANK
(4) RECYCLE TANK
(5) HUMIDIFIER
(6) NOx FEEDER
(7) VOLATILE RUTHENIUM GENERATOR
(8) OUTLET TREATMENT TOWER
(9) RUTHENIUM SAMPLER
(10) PUMP
(11) VACUUM PUMP
(12) FILTER
(13) MIST CATCHER
(14) HEATER
(15) THERMOCONTROLLER
(16) THERMOCOUPLE
(17) FLOW METER
(18) PRESSURE GAUGE
(19) MANOMETER
(20) DISTILLED WATER
(21) DRAIN
(22) AIR INTAKE
(23) STACK

Figure 2 Flow Sheet of the experimental apparatus.

The outlet of the scrubbing water is conected to the top of the scrubber with a pump in order to study the effect of scrubbing water recycle.

The volatile ruthenium was generated from the volatile ruthenium generator that contained non-radioactive ruthenium solution. The solution was prepared by dissolving RuO_4 in solution with sulfuric acid and potassium periodate. The air was fed into the flask with the solution and the RuO_4 was volatilized into the carrier gas. The carrier gas, the air with RuO_4, was flew into the main gas stream and the scrubber inlet gas concentration was controlled by adjusting the carrier gas flow rate. The RuO_4 volatility rate of the generator had been calibrated with the temperature and the carrier gas flow rate.

The ruthenium sampler consists of sampling bottles, a flow meter and a vacuum pump. The bottles contain 3(N) of hydrogen chloride and 5% ethanol solution. These samplers are installed at the gas inlet and the outlet pipe of the scrubber.

The experimental apparatus that contact with ruthenium are made of the glass to avoid the deposition of the ruthenium.

Procedures

Before the experiments the inside of the equipments was rinsed carefully to remove organic materials and to avoid the reduction of the RuO_4 on the surface of the glass. The experiments were performed as follows.

(1) The test gas without ruthenium was fed into the scrubber and the pump of the scrubbing water was started.

(2) The simulated off gas and the scrubbing water were kept on the temperature of the experimental condition. The temperature of the scrubber was also maintained by controlling the room temperature.

(3) The temperature of the scrubber inlet piping was raised to avoid deposition of RuO_2.

(4) The gas flow rate and scrubbing water flow rate were determined and confirmed to keep L/G.

(5) The carrier gas was fed into the ruthenium generator. The temperature and carrier gas flow rate were kept to attain an amount of the volatile ruthenium for the experiment.

(6) After the stable state of the experimental condition was attained, the gas sampling were started. The experimental and sampling time were determined to expect the concentration of the ruthenium in the sampling solution.

(7) The sampling bottles were separated from the experimental apparatus and the solution containing ruthenium was sent to the analytical laboratory.

(8) The DFs for the experiments were determined to calculate the ratio of the inlet ruthenium concentration to that of the outlet.

Experimental conditions

The experimental conditions were selected to simulate the actual operation range of the NOx absorbing scrubber that was installed in the HLLW vitrification off gas system. The concentration of the non-radioactive ruthenium was about 8×10^{-3} - 3×10^{-1} mg/Nm^3. The Ratio of the scrubbing water and gas flow rate were kept constant as L/G. The gas flow rate was determined to meet the linear gas velocity in the column. The flow rate of the scrubbing water was determined by gas flow rate and L/G. The standard temperature of the scrubber was 35°C. In addition to the standard experiment, the operational conditions were changed and these effects on the DFs were studied. Table 1 and Table 2 show the standard and various experimental conditions.

Table 1 Condition for the standard experiment

Feeding rate of scrubbing water	2.0 - 2.8[ℓ/h]
Mean gas temperature between the inlet and the outlet of the scrubber	33 - 37 [°C]
Temperature of scrubbing water at the inlet of the scrubber	36 - 38 [°C]

III Experimental Results

Standard condition

Figure 3 shows the results for the standard conditions together with the previous work. The figure denotes the relation of the volatile ruthenium concentration between at the inlet and outlet of the scrubber. The decontamination factor(DF) was calculated as follows.

$$DF = \frac{\text{Volatile Ruthenium Concentration of Inlet Gas}}{\text{Volatile Ruthenium Concentration of Outlet Gas}}$$

Table 2 Experiments for the various conditions

Experiment on low scrubbing water feeding rate;		
Feeding rate of scrubbing water	1.2	[ℓ/h]
Experiments on recycling of scrubber solution;		
Feeding rate of scrubbing water	0	[ℓ/h]
Recycling rate of scrubbing solution	2.2	[ℓ/h]
Experiment on high operating temperature;		
Mean gas temperature between the inlet and the outlet of the scrubber	47	[°C]
Temperature of scrubbing solution at the inlet of the scrubber	53	[°C]

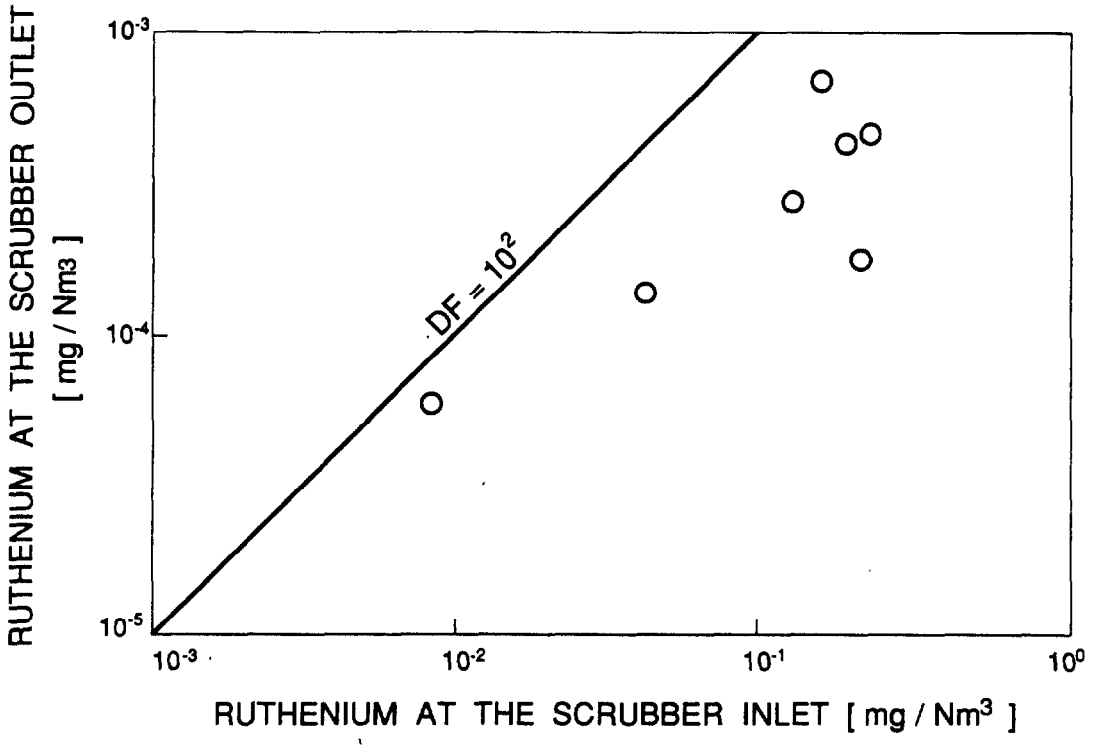
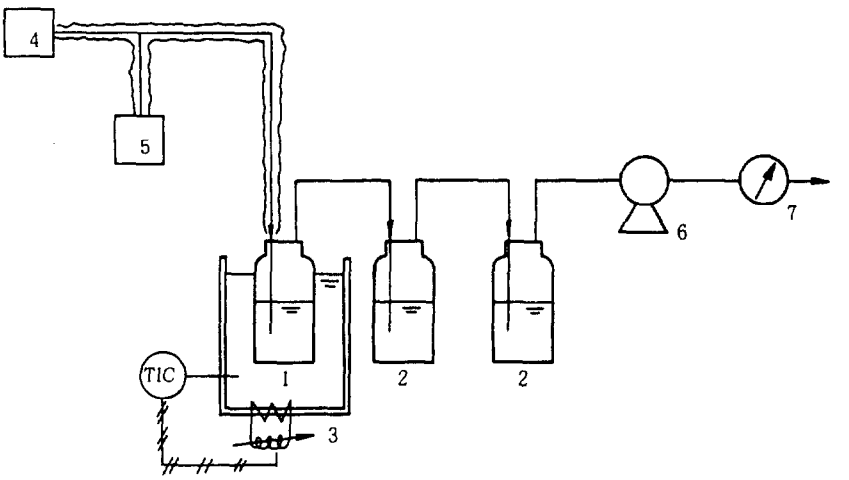


Figure 3 Relation between the scrubber inlet and outlet volatile ruthenium concentration; standard condition.

The straight line on the graph shows DF=100 and the result of the study shows the DF value of the scrubber is more than 100. It shows that the water scrubbing perforated plate column has high efficiency to remove the volatile ruthenium and the decontamination factor is comparable to that of the Silica-Gel adsorber.

Various conditions

The temperature of the scrubber may fluctuate as the results of the long operation. To evaluate the negative effect on the raising temperature, the basic test was carried out using a small absorbing bottle as Figure 4 before the test using the experimental scrubber. The ruthenium concentration of the test gas was 4.6mg/Nm^3. The result showed that the DF values are 40-60 for the temperature between 30°C and 50°C. The difference of temperature did not show the apparent effect on the DF.



(1) Absorbing bottle
(2) Sampling bottle
(3) Water bath
(4) Gas supply
(5) Volatile ruthenium generator
(6) Vacuum pump
(7) Gas flow meter

Figure 4 Equipment for the basic study.

The results of the various conditions in Table 2 are shown in Figure 5.

The test column, in Figure 2 was covered with the thermal insulator in addition to the control of the room temperature. The result at 47°C showed that DF value was kept more than 100 at this temperature. The result of the basic study was reconfirmed.

The flow rate of the scrubbing water was reduced to a half of the standard condition. The DF value was more than 100 as same as the result of the standard condition. The concentration of the ruthenium in the off gas and scrubbing solution were low enough not to affect on the DF value.

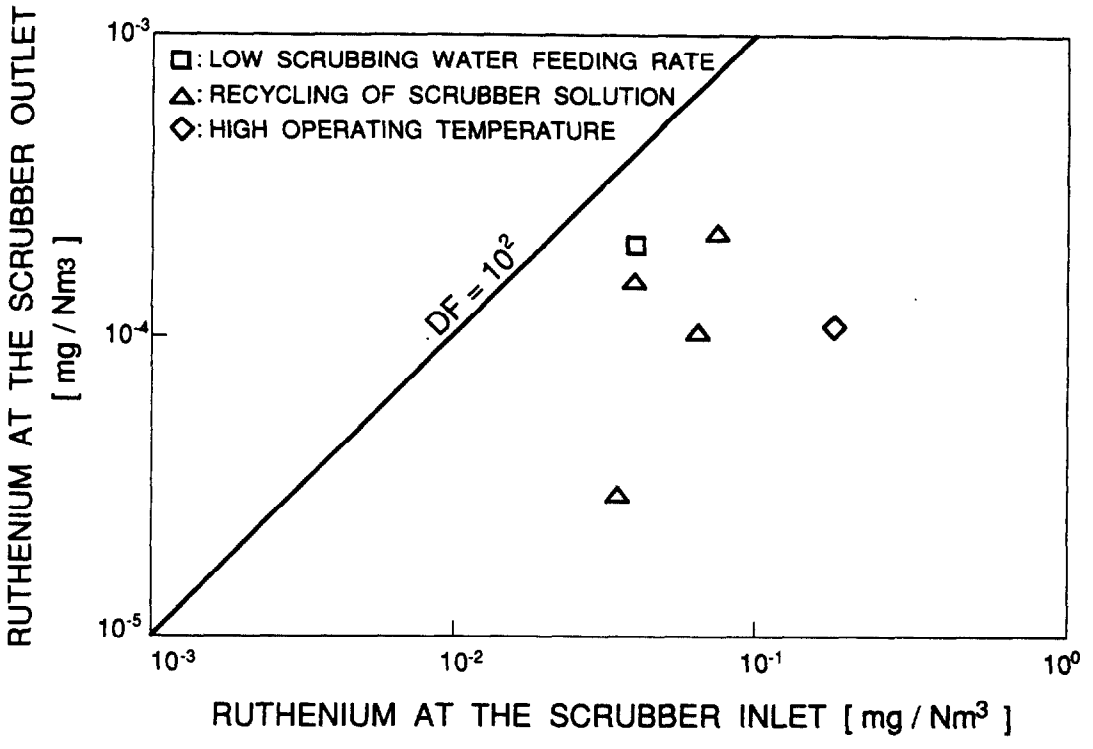


Figure 5 Relation between the scrubber inlet and outlet volatile ruthenium concentration; various conditions.

The scrubbing water was recycled to the top of the scrubber to simulate the various operation. The result did not differ from that of standard condition during two or three days operation. After the long term recycling operation, the scrubbing solution may be saturated by ruthenium and DF value may be decrease. But it takes very long period of operation because the ruthenium concentration of the gas is low and the increase of the concentration in the solution is very slow.

IV Conclusion

The experimental study on the volatile ruthenium decontamination factor of the perforated plate column scrubber was performed. The operating conditions were determined to simulate the condition at the NOx absorbing scrubber of the vitrification off gas process.

The decontamination factor for the standard condition was more than 100 and it did not decrease among various experimental conditions, raising temperature, reducing scrub water flow rate, and scrub water recycling. It is confirmed that the perforated plate column scrubber is an effective equipment to remove the volatile ruthenium. The decontamination factor for the volatile ruthenium of the vitrification off gas system with the perforated plate column is high enough to achieve nearly zero release rate of ruthenium.

References

1. Shirato, K., Kitamura, M., Asano, H. and Sakai, A., Ruthenium release rate from the pilot scale melter, in Proceedings of the Third International Conference on Nuclear Fuel Reprocessing Waste Management RECOD'91, Japan Atomic Industrial Forum, Tokyo Japan, Apr.1991, pp.781-785.

2. Shirato, K., Kitamura, M., Kanno, T., and Murakoshi, T., Experimental study on decontamination of volatile ruthenium with water scrubbing, in Proceedings of the 1989 Joint International Waste Management Conference, ASME, New York, NY, Oct. 1989, Volume 2, pp.223-226.

DISCUSSION

FURRER: I would like to give you a question. Have you observed any plateout of ruthenium or fallout of ruthenium on your scrubber plates? If so, you will have an increase of the dose rate on your plates. Can you answer the question?

KITAMURA: We have not observed plateout of ruthenium on the perforated plates. We kept the experimental temperature constant and we carefully and continuously scrubbed the inside of the plates to avoid the plateout of ruthenium.

MEASUREMENT OF CESIUM EMISSIONS DURING THE VITRIFICATION OF SIMULATED HIGH LEVEL RADIOACTIVE WASTE

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Abstract

In the Defense Waste Processing Facility at the Savannah River Site, it is desired to eliminate a startup test that would involve adding small amounts of radioactive cesium-137 to simulated high-level waste. In order to eliminate this test, a reliable method for measuring non-radioactive cesium in the offgas system from the glass melter is required. From a pilot scale melter system, offgas particulate samples were taken on filter paper media and analyzed by Inductively Coupled Plasma-Mass Spectrometry (ICP-MS). The ICP-MS method proved to be sufficiently sensitive to measure cesium quantities as low as 0.135 μg, with the sensitivity being limited by the background cesium present in the filter paper. Typical particulate loadings ranged from <0.2 to >800 μg of cesium. This sensitivity allowed determination of cesium decontamination factors for four of the five major components of the offgas system. The decontamination factors measured experimentally compared favorably with the process design basis values.

I. Introduction

The Defense Waste Processing Facility (DWPF) at the Savannah River Site (SRS) will be the United States' first facility to process high-level radioactive waste (HLW) into glass for storage in an off-site geologic repository. The Savannah River Technology Center (SRTC) operates the Integrated DWPF Melter System (IDMS), which is a pilot scale test facility used in support of the start-up and operation of the DWPF. Specifically, the IDMS is used in the evaluation of the DWPF melter and its associated feed preparation and offgas treatment systems.

The DWPF is currently preparing to begin its Cold Runs, in which non-radioactive, simulated waste materials will be used to test the operation of the facility. As part of the testing of the DWPF, several runs with simulated waste spiked with small amounts of radioactive cesium-137 are planned in order to determine the decontamination factors (DF) of the melter offgas system. Cesium-137 is the major radioisotope that is expected to enter the offgas system. Since the cesium-137 spike tests are to be the last tests conducted prior to radioactive startup, it was desired to determine if the decontamination factors for cesium removal could be ascertained in the initial Cold Runs so that any equipment or processing modifications could be implemented as soon as possible. Moreover, substantial cost and time savings could be realized if the cesium-137 spike runs could be eliminated.

An experimental program was conducted in the IDMS to demonstrate the feasibility of determining cesium decontamination factors with non-radioactive cesium since non-radioactive cesium analysis techniques were believed to lack the required sensitivity. In addition to measuring the DF for cesium, the DF for total particulate was also measured.

A. IDMS Process Description[1]

An overall flow diagram of the IDMS melter and offgas system is shown in Figure 1. The glass melter in the IDMS is a joule-heated slurry fed melter which melts glass at 1050-1100 °C. A slurry of HLW and borosilicate glass frit is fed via a feed tube onto the top of the glass surface in the melter. The melter is equipped with Inconel resistance heaters in the vapor space to assist in vaporization of the water in the slurry feed and to combust offgases evolved from the slurry. The melter is purged with air and the offgas passes through a film

Work performed under U. S. Department of Energy Contract No. DE-AC09-89SR18035

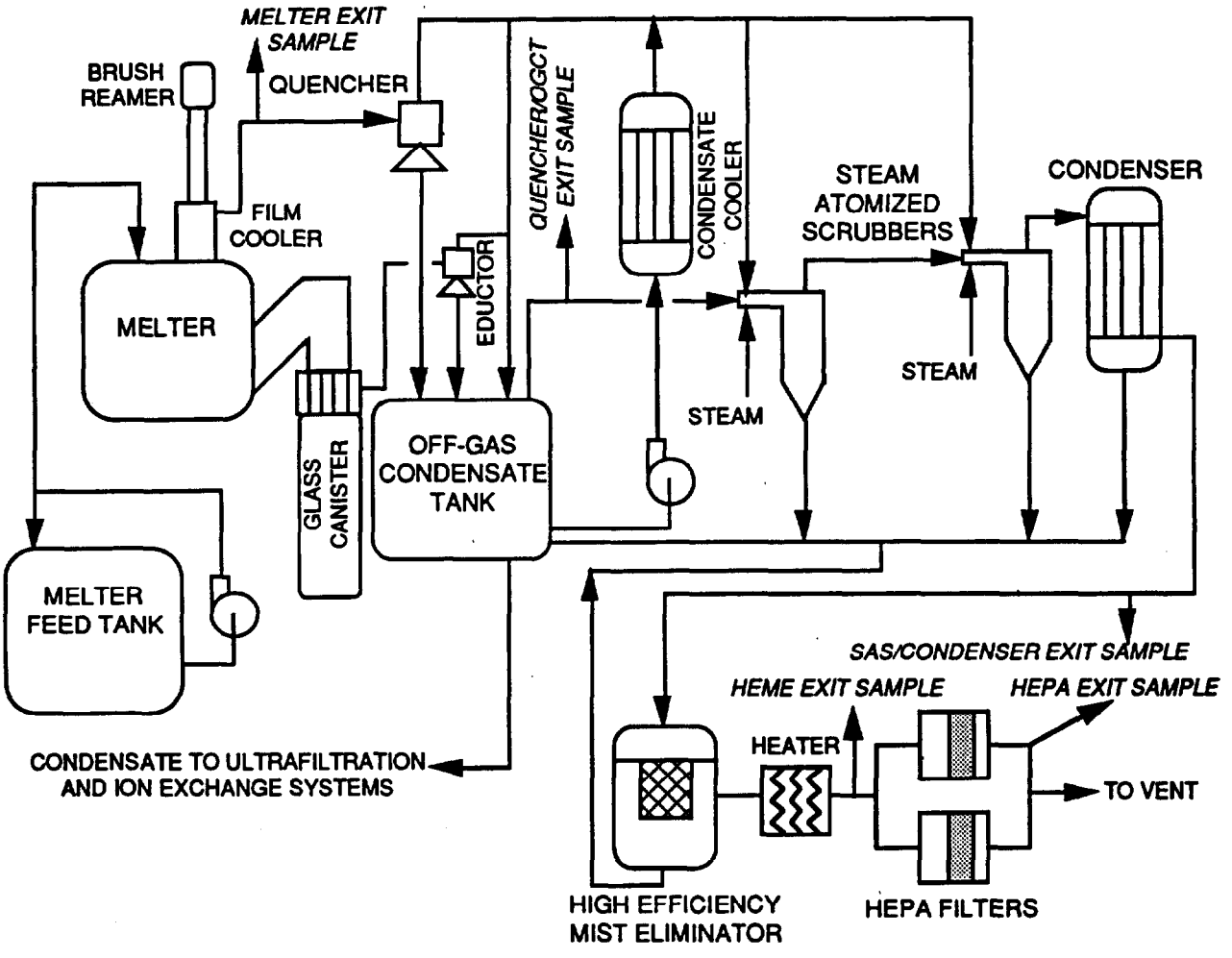

Figure 1. Integrated DWPF Melter System offgas system schematic.

cooler designed to minimize particulate build up in the offgas line and to cool the offgas from 450-725°C to less than 350°C.

After passing through the film cooler, the gas is then scrubbed in the Quencher (Figure 2), which is an ejector-venturi scrubber that reduces the gas temperature below the dew point to disengage most of the water vapor from the non-condensibles, scrub entrained glass particles, and allow semi-volatile particles (salts) to coalesce. The Quencher uses offgas condensate as the motive fluid. The offgas and condensate leaving the Quencher enter the Off-Gas Condensate Tank (OGCT) where the liquid and vapor disengage. The condensate is maintained at 40°C by a cooler.

The offgas from the OGCT is then passed through a series of two Steam Atomized Scrubbers (SAS)[2], which remove submicron and micron-sized particles (Figure 3). The SAS removes particulates by combining water with the offgas in a region of turbulent mixing. This mixing action is created by directing a jet of steam into an enclosed cavity so as to create a region of turbulent flow along the outer boundary of the jet. Water is sprayed around the jet and mixes with the process gas. The droplets formed are then accelerated in the mixing tube where turbulence is sufficient to promote coalescence, but low enough so as not to cause significant re-entrainment of the liquid. The droplets are separated from

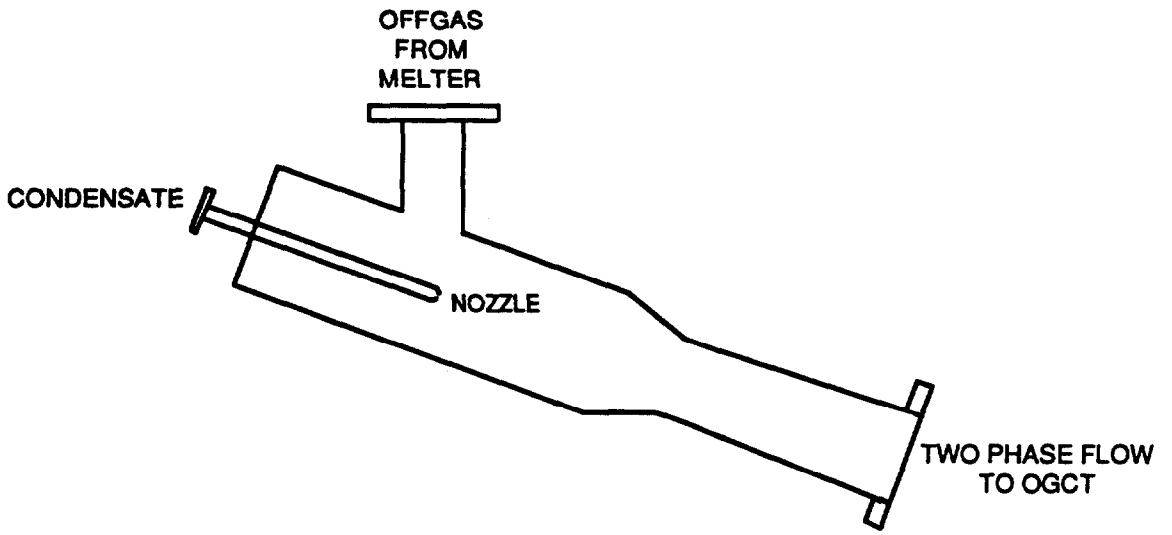

Figure 2. Melter offgas system Quencher.

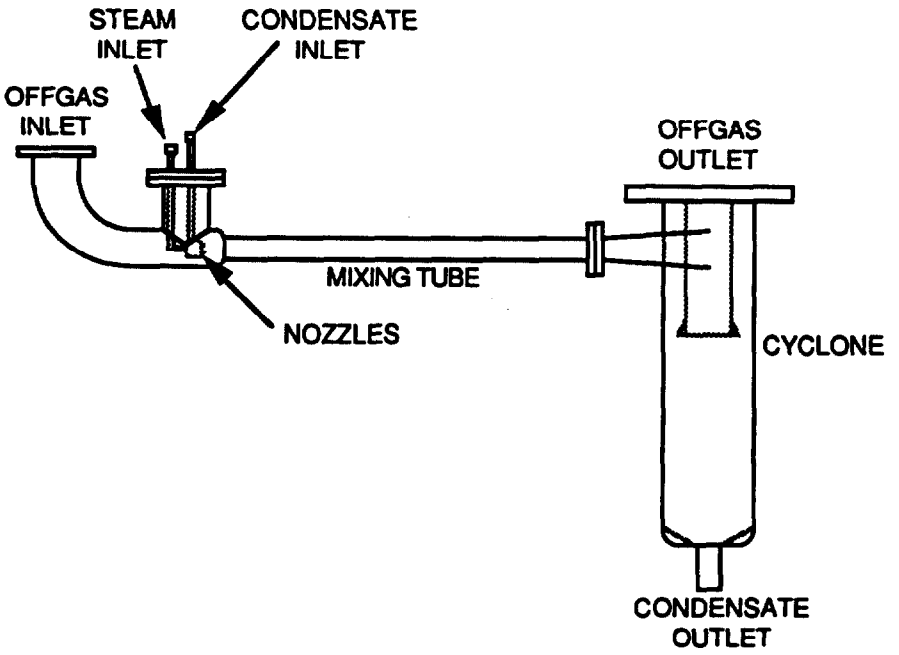

Figure 3. Melter offgas system Steam Atomized Scrubbers.

the vapor in a cyclone separator. The SAS technology was developed and patented by the Lone Star Steel Company, which refers to the equipment as Hydro-Sonic Scrubbers. The condensate and condensed steam are returned to the OGCT.

The offgas leaving the SASes is passed through a shell-and-tube heat exchanger that separates the condensibles from the offgas and removes virtually all of the elemental mercury. The separated condensibles are returned to the OGCT. A mist eliminator (York

demister) with an atomized water spray removes any suspended liquid droplets from the non-condensible gases.

The High Efficiency Mist Eliminator (HEME) consists of a densely packed glass fiber filter that is wetted continuously by an atomized water spray. There is a coarse 1.25 cm layer of 30 μm glass fiber on the face followed by a 6.4 cm layer of fine 8 μm glass fiber packed at 175 kg/m^3. The HEME was designed to operate at a 2.5 cm/sec superficial face velocity.

After passing through the HEME, the offgas is heated 10°C above its dew point to prevent condensation in the High Efficiency Particulate Air (HEPA) filters. Two HEPA filters in parallel serve as the final treatment step before the air is released to the environment. In DWPF, two HEPA filters are used in series, followed by a sand filter.

II. Experimental Methods

A. Sampling

The particulate samples were taken from five different points in the melter offgas system. The locations of the sampling points within the process are shown in Figure 1. The five sample points are identified as:

1. Melter Exit
2. Quencher/OGCT Exit
3. SAS/Condenser Exit
4. HEME Exit
5. HEPA Exit

A modified version of the EPA Method 5[3] Sampling Train was used to sample the IDMS melter offgas system at the five locations. The equipment used is shown in Figure 4. The offgas was sampled at as near to isokinetic conditions as possible. The process mass flowrate was measured by process flow transmitters and converted to velocity and corrected for variations in pressure, temperature and density. The Staksamplr (Anderson Samplers) gas metering system was adjusted in real time to keep the sample velocity equal to the process velocity. Particulate samples were collected on glass fiber filter paper (Reeve Angel 934AH Mark 3/5 #1) with an average pore size of 0.3 μm. After leaving the filter housing, the sampled offgas passed through four (or five) chilled (0-5°C) impingers which condensed water and other condensible gases.

As a check of the accuracy of sampling, the approach to 100% isokinetic sampling (%I) was calculated for all samples. The desired %I is 100%, with 90-110% being acceptable to the EPA for pollution monitoring. Experience with another melter system at SRTC has shown that it is difficult to keep the % isokinetic in this range for pipes of 5-10 cm diameter. EPA Method 5 is designed for sampling of stack gases in which the velocity does not vary significantly in time. However, for the melter system, the offgas velocity can vary substantially over the time intervals sampled. To calculate the %I, time averaged process temperatures, pressures, densities and flowrates were used. Hence, the actual %I will be better than that calculated from the time averaged data. For sampling of IDMS, the sample weighted average %I was 94% with a standard deviation of 18%; the maximum was 132% (1 sample) and the minimum was 68% (1 sample). Four samples were taken at a %I of 200%; these samples were not considered in the calculations of the decontamination factors.

B. Analyses[4]

The filter samples containing the collected IDMS offgas line particulates were sent to Lawrence Livermore National Laboratory (LLNL) for examination by ICP-MS. First, x-ray

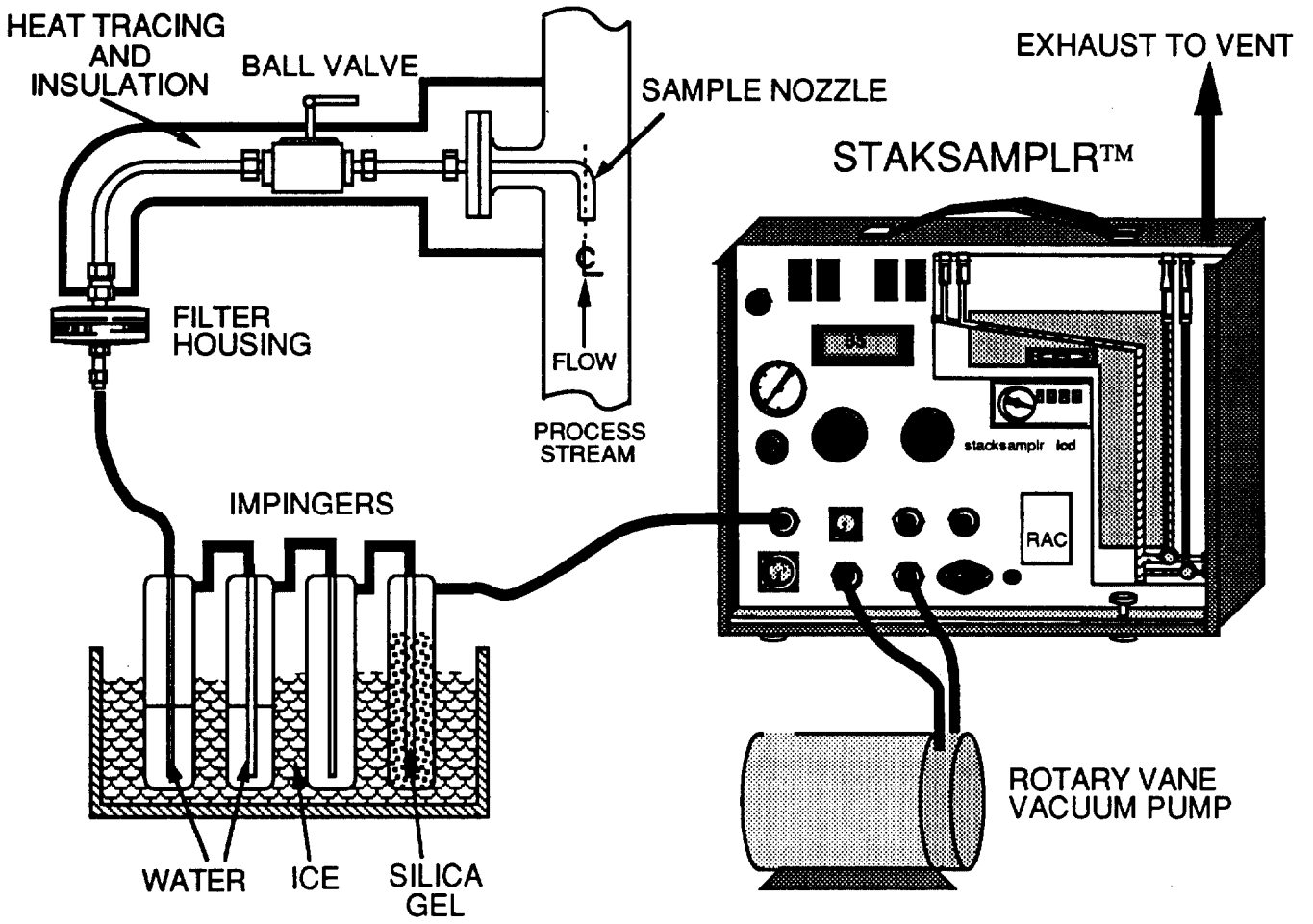

Figure 4. Modified EPA Method 5 sampling train.

fluorescence was used to obtain an approximate value for the cesium present in each sample. The samples were then dissolved in a mixture of nitric and hydrofluoric acids, taken to dryness, and redissolved in dilute nitric acid. Additional dilutions were made to bring the cesium concentrations to levels matching the instrument's range of optimum performance. Only two dilution levels, differing by a factor of ten, were used. The total cesium per sample ranged from 0.34 to 19.2 μg and 9.18 to 948 μg for the smaller and larger dilutions, respectively.

Three blanks were used in the determination: nitric acid, reagent, and filter medium. The amount of cesium in the nitric acid used for the dilutions was first determined. The reagent blank, which included all the materials used, was measured at each of the two dilution levels. Two filter medium blanks, one at each dilution level, were also dissolved, analyzed, and reanalyzed. As expected, the filter medium blanks showed the highest amount of cesium and limited the sensitivity of the analyses. The filter medium blanks contained an average of 0.315 and 0.357 μg cesium for the low and high dilution, respectively; these values were subtracted from all of the measured cesium values.

After dissolution, roughly half of the samples analyzed left insoluble residues which were centrifuged out. The residue is believed to be associated with the filter paper medium, since some of the blanks also gave a residue. In all but one sample, the residue contained less than 1% of the total cesium; in that one sample, the residue contained >31% of the cesium.

To ensure the integrity of the analyses, several original solutions were re-diluted and reanalyzed. In addition, two blanks including filter papers were spiked with 10 and 100 µg of cesium. Three standards solutions (1, 10, 100 ppb) were also analyzed and a 10 ppb solution was analyzed before and after each sample to correct for drift. The calibration curve of the ICP-MS was linear and drift was minor. The uncertainty of the ICP-MS technique, based on duplicate analyses, ranged from 0.0 to 7.1%, with an average uncertainty of 4.4%.

Total particulate weight was determined by weighing the filter papers on a 5 place analytical balance before and after sampling. For both the initial and final weight, the filter papers were dried in an oven at 110°C for at least 24 hours and were cooled in a desiccator. In some cases, the weight of the filter paper after sampling was so close to the initial weight that the DF determined was very inaccurate.

III. Results and Discussion

Cesium concentrations greater than the blanks were obtained from four of the five locations sampled (Table 1). The HEPA exit location gave samples that were only slightly higher than the blanks, with substantial variation in the weights for a given sampling interval. The HEME exit samples were also only slightly larger than the blanks, but the weights were more consistent. If a filter paper with less background cesium were available, the HEME exit measurements would be more accurate, but the HEPA exit measurements would probably not be improved. The typical sampling times used for each of the sampling locations are shown in Table 2. Greater sampling times would increase the cesium loading on the filter paper, but times greater than 10 hours were not possible because the filter papers would often become damp and result in a pressure drop too high to continue sampling. SRTC is presently evaluating a method in which the offgas samples are scrubbed with ultrapure nitric acid to collect the cesium; no filter paper is used in this method.

Table 1. Amounts of cesium measured on filter paper samples.

	Weight (µg)		
Location	Lowest	Mean	Highest
Melter Exit	8.82	209.54	445.64
Quencher/OGCT Exit	87.04	348.65	862.64
SAS/Condenser Exit	1.815	23.90	132.64
HEME Exit	0.135	0.338	0.775
HEPA Exit	0.025	0.245	0.705

Table 2. Typical total sampling times.

Location	Time
Melter Exit	4 - 15 min
Quencher/OGCT Exit	1.5 - 4 hr
SAS/Condenser Exit	1.2 - 4 hr
HEME Exit	4 - 8 hr
HEPA Exit	4 - 10 hr

Table 3 shows the cesium flowrates measured in the IDMS offgas system. This table also shows the design decontamination factors for the DWPF and the overall

decontamination factors measured in IDMS. Comparison of the overall decontamination factor (without the HEPA filters) shows that IDMS achieved an order of magnitude better performance than the design basis, mainly due to the high DF for the melter. The IDMS melter has a much larger vapor space volume to melt surface area ratio than the DWPF melter which may result in less entrainment of glass particles.

Table 3. Design and measured cesium decontamination factors.[5]

DF across:	IDMS Cesium Flowrate (mg/min)	DWPF Design DF	IDMS Measured DF
Melter Feed	335.	-	-
Melter	2.54 ± 1.39	15	132
Quencher/OGCT	0.960 ± 0.560	8.9	2.7
SAS/Condenser	0.0337 ± 0.0208	50	28.5
HEME	$1.07 \pm 0.78 \times 10^{-4}$	40	314
HEPA #1	$1.43 \pm 1.91 \times 10^{-4}$	422	0.75
HEPA #2	NA	24.5	NA
Overall (without HEPAs)	-	2.67×10^{5}	31.3×10^{5}
Overall (without HEPAs & feed to offgas)		1.78×10^{4}	2.37×10^{4}

The DF for the Quencher/OGCT in IDMS was much lower than the design basis. It is reasonable to assume that the concentration of larger particulates at the inlet to the Quencher was smaller than assumed in the design basis since the DF of the melter was much higher than expected. The Quencher/OGCT combination is designed to remove large particulates, which are mainly entrained glass,[6] so it is not unexpected that it performed less efficiently than the design basis. Table 4 shows the design basis decontamination factors for both entrained and semi-volatile cesium. The SAS/Condenser DF was only about 56% of the design value; however, within experimental error, this equipment probably performed up to the design basis. The HEME filter DF indicates that this filter has an overall efficiency that is much better than that assumed in the design.

The total particulate decontamination factors determined in IDMS and the DWPF design values are shown in Table 5. The overall DF determined in IDMS is similar to the design value, but its magnitude is mainly due to the melter DF being much larger than expected. Again, the low experimental decontamination factors are due to there being less entrained particulate in offgas (melter DF = 38 times design) and also due to the difficulty in measuring small weight gains on the filter papers.

Table 4. Design decontamination factors for entrained and semi-volatile Cesium.

DF across:	Entrained Cesium	Semi-volatile Cesium	Overall Cesium
Quencher	10	1	8.9
SAS/Condenser	50	50	50
HEME	40	40	40
HEPA #1	500	200	422
HEPA #2	50	10	24.5

Table 5. Design and measured total particulate decontamination factors.

DF across:	DWPF Design* DF	IDMS Measured DF
Melter	69	2626
Quencher/OGCT	<10	2.85
SAS/Condenser	50	20.6
HEME	40	8.2
HEPA #1	<500	1.43
HEPA #2	<50	NA
Overall (without HEPAs)	$\approx 1.38 \times 10^6$	1.24×10^6

* design DF values indicated are for entrainment only; semi-volatile values will be smaller

IV. Conclusions

The data indicate that the decontamination factors for cesium in the Melter, Quencher/OGCT, SAS/Condenser and HEME can be measured by sampling the offgas using standard filter paper sampling techniques. The amount of cesium collected at each location is sufficient to make accurate measurements, although the HEME Exit location would benefit from the use of a filter paper medium containing less background cesium. Measurement of cesium at the HEPA Exits is not possible with this technique. For both the HEME and HEPA Exit sample locations, the nitric acid scrubber method may be feasible, but experimental results are needed before this method can be recommended.

Based on the results reported here, and assuming that the nitric acid scrubber method works for the HEME and HEPA exit locations, the decontamination factors in the DWPF can be determined without the need to spike the melter feed with cesium-137.

The decontamination factors determined in IDMS indicate that the DWPF melter offgas system components should perform at or above their design specifications for the removal of cesium.

Acknowledgements

The authors would like to acknowledge the assistance of: Price Russ (LLNL) for his analyses; Stacey Adamson (WSRC) in issuing the initial report; and Frances Williams, Glenn Parker, Richard Creech and Larry Bozard (WSRC) for countless hours of sampling.

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DISCUSSION

FIRST: The later slides you showed with the decontamination factors in the train, suggest to me that you have too much air cleaning equipment in there; far more than you need because you are not really exercising all the equipment. Having more than you need is a safety factor, but what is bad about it is that you take all of your contamination and divide it up into so many little different packages that you increase your disposal problem.

ZAMECNIK: Let me show the slides again and explain it a little better. The way the off-gas system is set up the scrubbing solution that we use in all the scrubbers is the condensate from the melter. The off-gas is about 50% water and so we are continuously forming condensate that is recirculated through the cooler and is used in the quench water coming back to the condensate tank. It also is used in both scrubbers and then returns to the condensate tank. Condensate from the HEME also returns back to the condensate tank. So we are collecting as much of the contaminants as possible in one spot. Ultimately, in the IDMS pilot plant, we send it to a waste treatment system. But in DWPF, it will be send back to the tank farm and to the evaporators and it will come around again in a future melter feed. The only things in this system that we have to dispose of, that don't end up going in the condensate, are the HEME pads in the HEME, and the HEPA filters. Since these are both glass-fiber filters, plans are to have a system in DWPF which will take the filters, dissolve them in caustic, add it to the feed preparation system, and recycle it back through the entire vitrification plant. So, our only effluent from the entire facility is the final scrubbed off-gas.

BERGMAN: Why does the second HEPA filter have one tenth the decontamination factor of the first, or ten times higher penetration?

ZAMECNIK: The DF for the first HEPA is smaller than would normally be assumed since the upstream scrubbing equipment (Quencher, Steam Atmosized Scrubbers, HEME) will have removed a significant amount of the particulate. Thus, the HEPA is presented with particles that are predominantly around 1 μ or less; thus, we assumed a DF of only 500. Similarly, for the second HEPA in series, a DF of only 50 was assumed since the particulates presented to it will be much more difficult to remove.

GOOSSENS: I would like to ask if you are planning long duration tests? Can you perhaps explain what will be the duration of these tests; weeks or months?

ZAMECNIK: Two test were made over the 3rd and 4th experimental runs. Each set of samplings was about two weeks. The number of particulate samples that went into calculating each of the DFs was, on average, 30 - 40. As of now, in the pilot-scale facility, we do not plan further tests. DWPF will be doing extensive testing throughout the two year period of the cold runs.

22nd DOE/NRC NUCLEAR AIR CLEANING AND TREATMENT CONFERENCE

STRUCTURAL TESTING OF SALT LOADED HEPA FILTERS FOR WIPP

by
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Abstract

The ventilation studies of the Waste Isolation Pilot Plant described in this paper were performed by personnel from New Mexico State University in collaboration with Sandia National Laboratories, Los Alamos National Laboratory and Westinghouse Corporation. High efficiency particulate air filters (0.61m by 0.61m by 0.3m) of the type in use at the Waste Isolation Pilot Plant were loaded with salt aerosol provided from that site. The structural strength of salt-loaded, high-efficiency filters was investigated at two humidity levels, high (75%RH) and low (13-14%RH), by subjecting the filters to pressure transients of the types expected from tornadoes. Filters loaded under the high humidity condition proved to have a greater structural strength than did the filters loaded under the low humidity conditions, when both types were subjected to tornado-like pressure pulses. This unexpected result was apparently due to the crystallization of salt upon the wire face guard of the HEPA filter loaded under the high humidity condition which kept salt from penetrating the filter medium while still providing a substantial pressure drop at the standard flow rate. Results are also presented for HEPA filters pre-conditioned at 100%RH before structural testing and for HEPA filters in series with pre-filters.

I. Introduction

The Waste Isolation Pilot Plant (WIPP) near Carlsbad, New Mexico is an underground repository designed for the safe disposal of transuranic wastes. Approximately 14.5 kilometers of horizontal shafts have been excavated in a stable salt formation 660 meters below the surface. Continual activity within the repository generates a salt aerosol that is easily moved by the ventilation system of the facility. In a previous paper[1] we reported on the affect of this salt aerosol upon the filtering efficiency and flow resistance of the WIPP site High Efficiency Particulate Air (HEPA) filters. In this paper we examine the affect of salt loading upon the structural strength of HEPA filters when subjected to tornado-like pressure transients and shock overpressures. Two humidity levels were chosen, 13-14%RH, which is well below the point at which salt acquiesces, and 73%RH, which is well above this point.

II. Review of Methodology

Salt Loading of HEPA Filters

The experimental determination of HEPA filter structural strength as a function of salt dust loading was carried out in the Large Scale Flow Facility (LSFF) of the Mechanical Engineering Department at New Mexico State University. Figure 1 is a schematic diagram of the apparatus upon which clean HEPA filters from the WIPP site were loaded with naturally occurring salt dust from WIPP site tunnels. These filters were standard 0.61m by 0.61m by 0.3m in size and had been certified by the Rocky Flats test facility to have pressure drops of no more than 2.54cm of water at the rated flow of $28m^3/min$ and efficiencies of at least 99.97%. See Ref. 1 for details on loading.

Structural Strength Testing of Salt Loaded HEPA Filters

The structural strength of salt loaded HEPA filters was investigated for pressure transients which simulated both tornado-like and explosive-like pulses.

Tornado Testing: Simulation of tornado-like pressure pulses was carried out in the blow-down wind tunnel shown in schematic in Fig. 2. This wind tunnel consisted of a 0.61m by 0.61m square duct connected to a 3.05m by 3.05m by 3.05m plenum chamber. Two large high pressure tanks supplied the plenum chamber through 12 sonically limited solenoid valves. The valves were computer controlled to produce a pressure pulse across the test HEPA filter which closely approximated the NRC Region I tornado pressure signature (see Fig. 3). The downstream face of the 0.61m by 0.61m by 0.3m HEPA filter being tested was photographed by a Red Lake high speed camera at 100 frames per second. The camera was synchronized, through a PDP 11/73 computer, with the pressure drop across the filter in order to determine the pressure difference which caused structural failure of the filter. The point of filter failure was defined as that instant when the first crack or tear appeared in the filter media.

Shock Testing: Simulation of explosive-like shock waves was accomplished in a 0.91m diameter shock tube shown in schematic in Fig. 4. The shock tube driver section (high pressure section) was 3m in length, and the driven section (low pressure section) was 36.6m in length. The test HEPA filter was located at the outlet end of the shock tube (far left in Fig. 4). The Red Lake high speed camera was used to photograph the downstream face of the test filer at 500 frames per second during the passage of the shock wave through the filter in order to determine the instant of filter failure. The camera was again synchronized with the pressure difference across the filter through the PDP 11/73 computer. Figure 5 shows a typical pressure signal of a shock wave approaching the test filter.

Data Acquisition: The PDP 11/73 computer was coupled with a CAMAC data acquisition system capable of data rates as high as 1Mhz on 24 channels. Normally, data was taken at slower rates to conserve memory and the highest rate was used only in the case of shock wave passage,

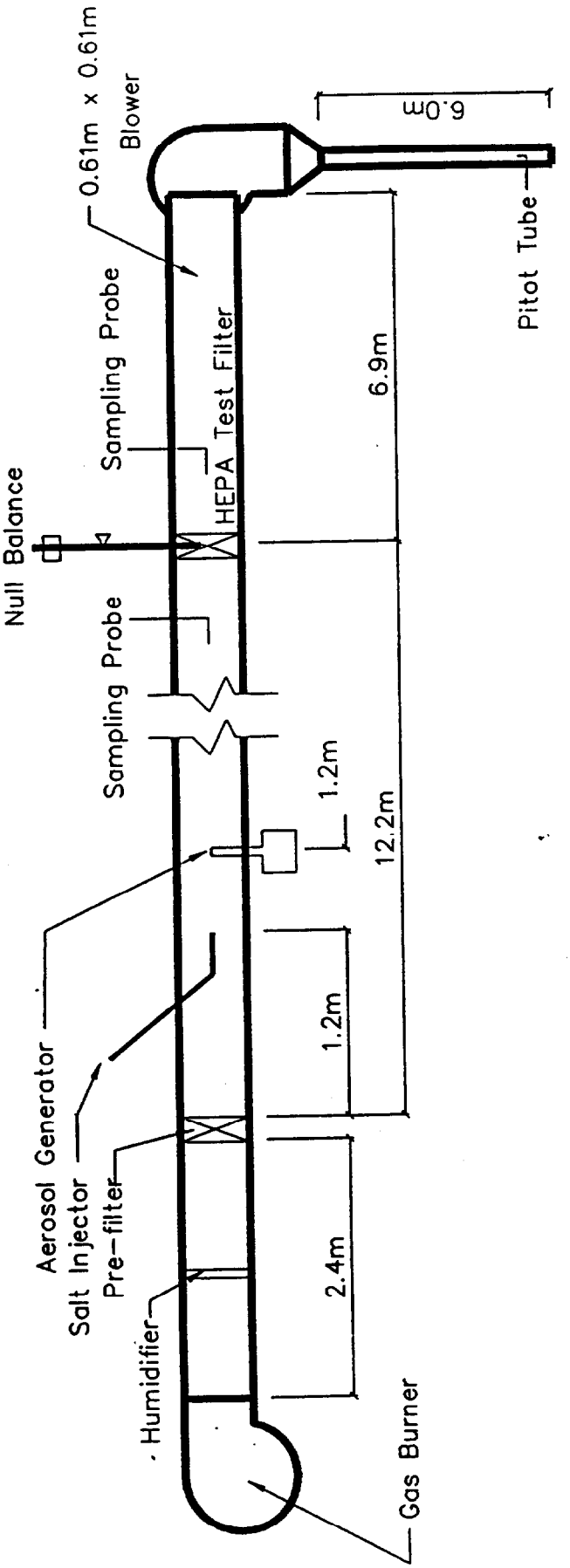

Figure 1 HEPA Filter Aerosol Salt Loading Apparatus

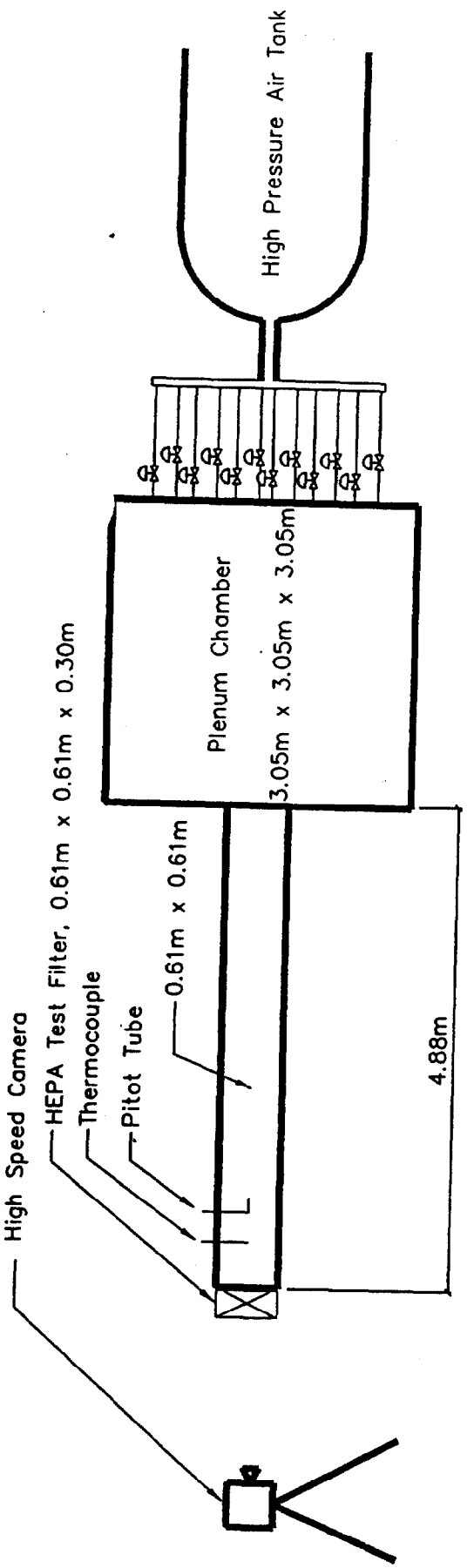

Figure 2 Blow-Down Wind Tunnel Tornado Pressure Pulse Simulator

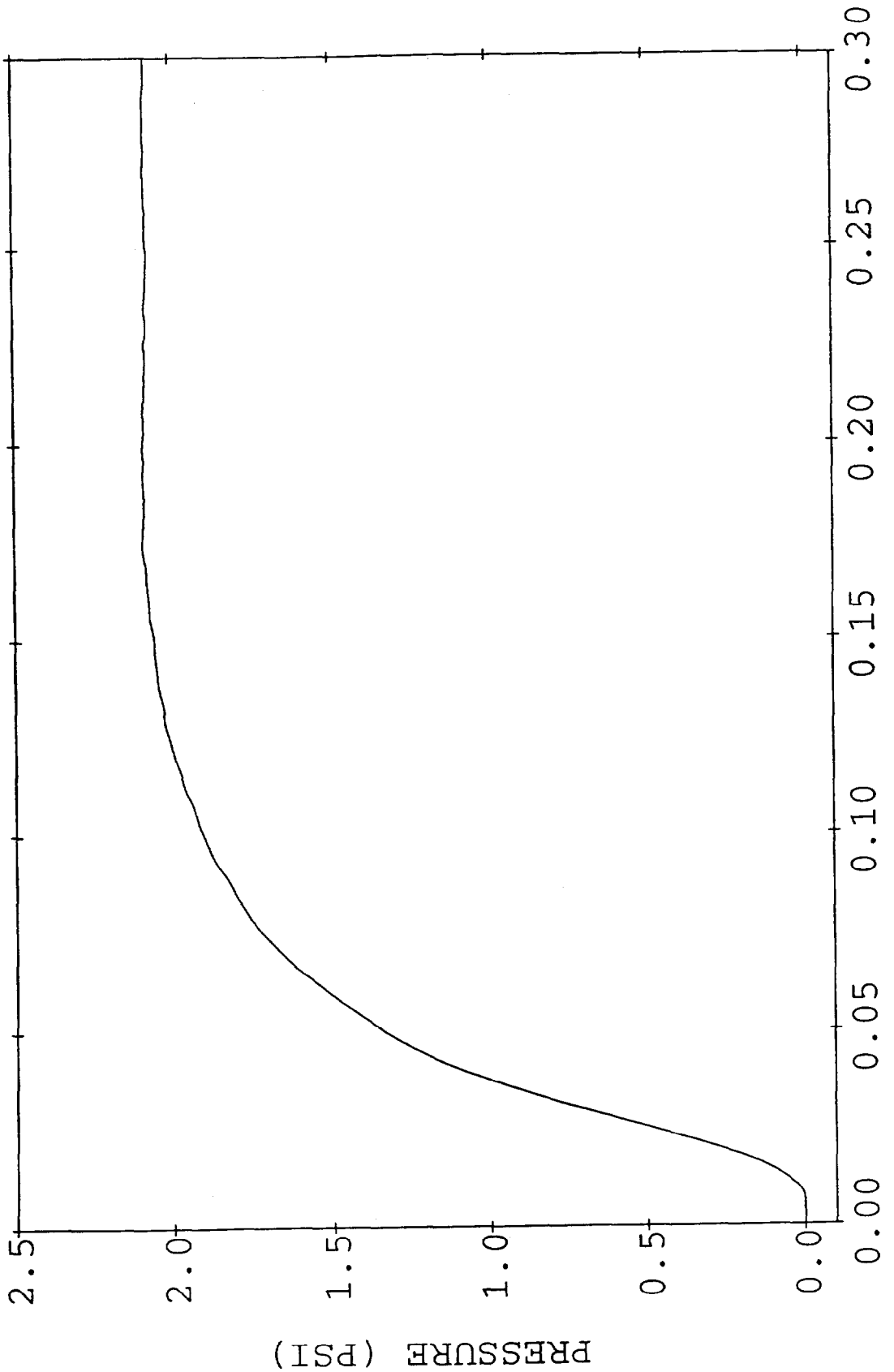

Figure 3 Approximation to Region I Pressure Pulse Across HEPA Filter in Tornado Simulator.

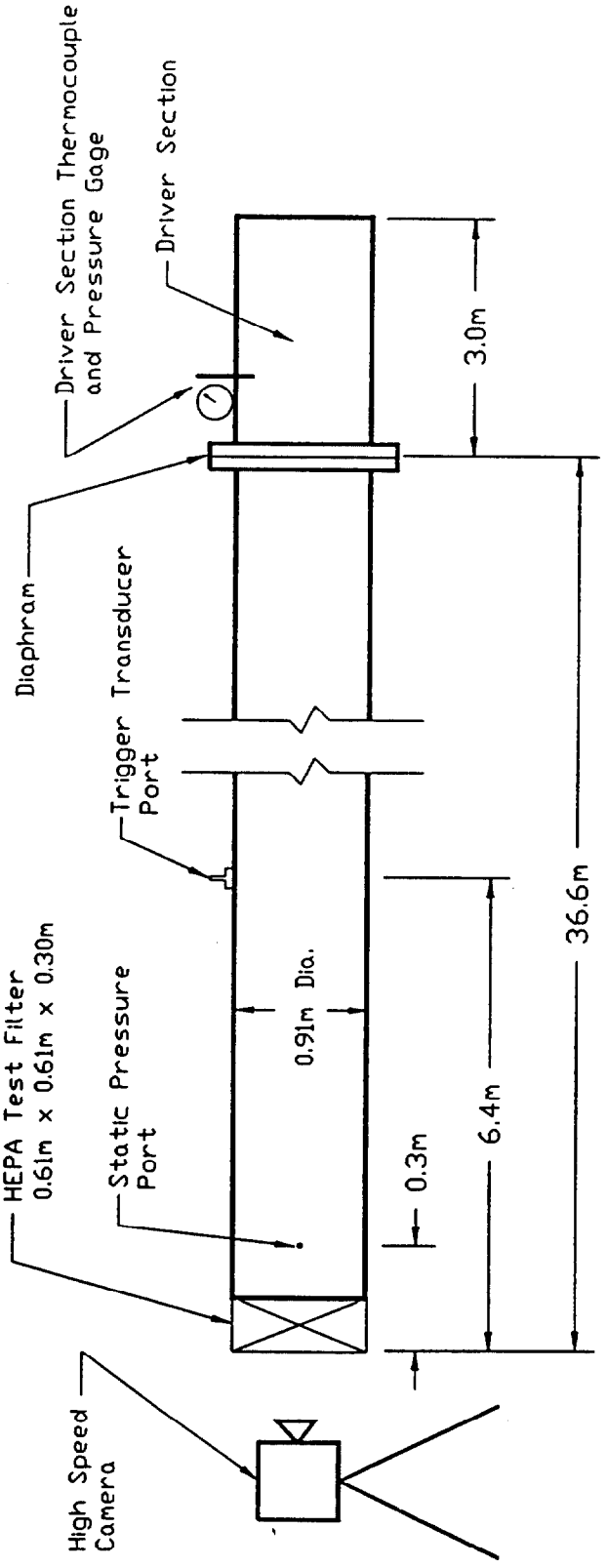

Figure 4 Shock Tube Explosive Pressure Wave Simulator

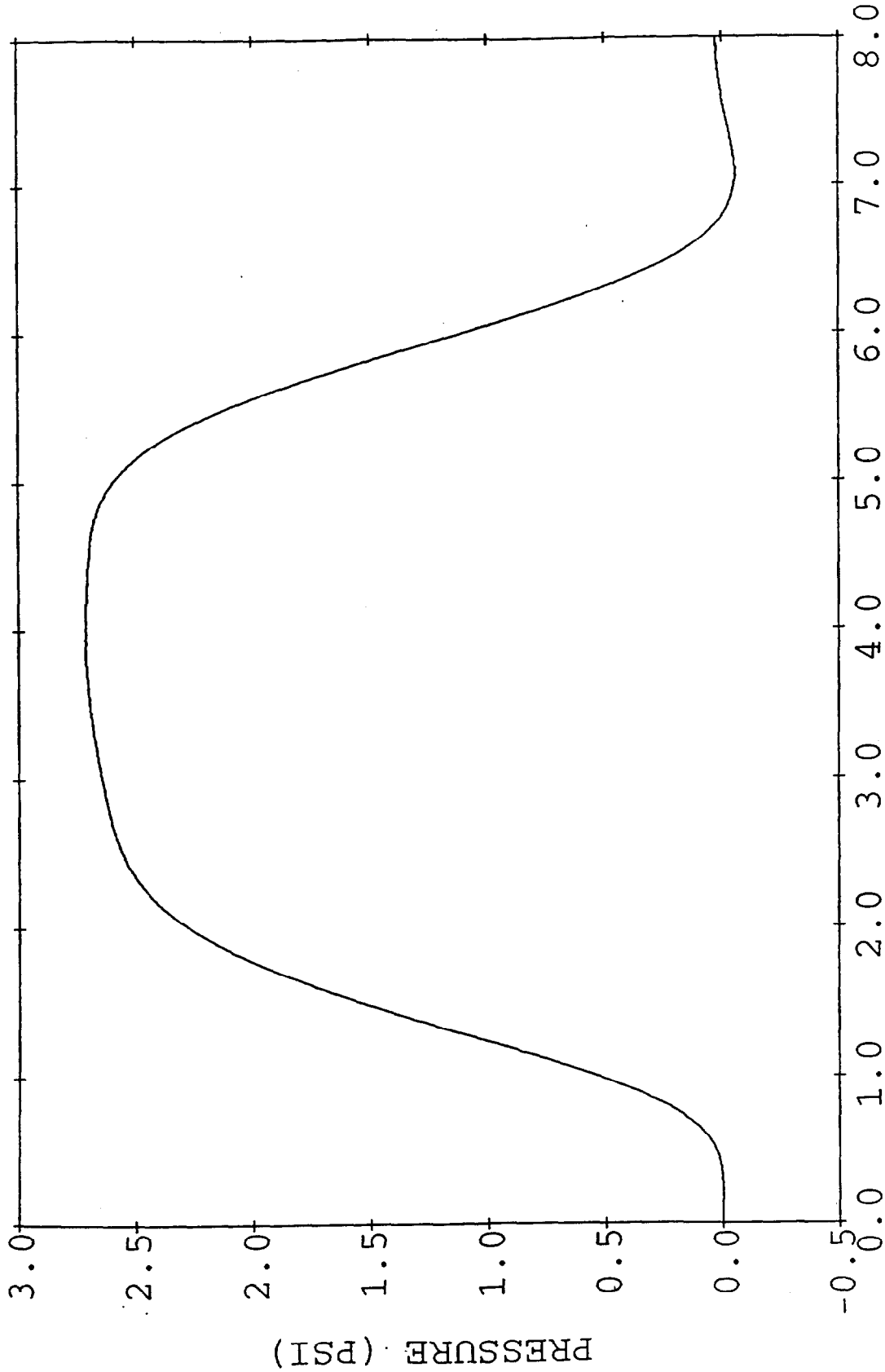

Figure 5 Typical Shock Overpressure at HEPA Filter in Explosive Wave Simulator.

since the leading edge of the shock wave was on the order of 50μsec in width.

III. Discussion of Results

The following summarizes the results of the structural testing of salt loaded HEPA filters.

Structural Response of HEPA Filters to Pressure Transients

Experiments to determine the structural strength of salt loaded HEPA filters were carried out in the tornado simulating blow-down wind tunnel and the 0.91m diameter shock tube. Filters were tested under high and low humidity conditions in the tornado simulation experiments.

Tornado Tests: Table 1 summarizes the results of the HEPA filter tornado blow down tests which were performed. Six tests were run: Two filters loaded with salt under low humidity conditions (approximately 13%RH and 14%RH); Two filters loaded with salt under high humidity conditions (approximately 73%RH and 84%RH); a single clean, unloaded HEPA filter; and a clean, unloaded HEPA filter in series with a clean, unloaded pre-filter. The break pressures at which first failure of the HEPA filter medium was detected is shown in Table 1. The maximum expected error of the break pressures is 0.26psi (1.8kPa). Notice, for single filters, this pressure drop ranged from 0.504psig (3.46kPa) for filter SN184783 to 1.226psig (8.42kPa) for filter SN185234. The two clean, unloaded filters (HEPA SN184678) in series failed at a pressure drop of 2.052psig (14.1kPa) across the combined filters. All the salt loaded filters were originally loaded to at least 3 inches (7.62cm) of water pressure drop at 1000cfm before undergoing structural testing, because at the WIPP site filters are changed out when they reach this level of loading. The filters which were loaded with salt under controlled humidity conditions usually were tested structurally in the tornado blow down facility immediately upon the completion of the salt loading process so that the combined effects of the trapped salt and moisture would be evident. However, one filter (SN184783) was conditioned with a flow having 100%RH for four hours before structural testing and the filter medium of this filter became saturated with water. All the filters tested for structural strength were standard 0.61m by 0.61m by 0.3m HEPA filters and were provided to us from the WIPP site.

Figures 6 through 11 show the recorded pressure differences across the HEPA filters during the tornado blow down tests. Figure 12 is a composite of Figs. 6 through 11 showing all the pressure signatures on one plot for comparison. Notice that all the pressure signatures are different than the expected Region I tornado induced pressure difference shown in Fig. 3. This is because each filter failed catastrophically (i.e. the entire filter pack was torn out of its frame) before the desired peak pressure was reached. However, notice for all six filter tests that the initial slope of the pressure difference-time curve was very close to the desired pressurization rate shown in Fig. 3.

Table 1 HEPA Filter Tornado Blow Down Tests.

Filter Condition	Serial No.	Break Time (sec)	Break Press. (psig)	Peak Press. (psig)
Low Humidity Filter	185274	0.97	0.680	0.868
Low Humidity Filter*	184659	1.21	0.843	0.932
High Humidity Filter*	184684	1.28	1.180	1.228
High Humidity Filter+*	184783	1.16	0.504	0.519
Clean Filter	185234	1.42	1.226	1.248
Clean/Series Filters	184678	1.49	2.052	2.226

+Filter conditioned at 100% R.H. for 4 hrs. before structural test.
*Maximum expected error may be somewhat larger than reported.

Table 2 HEPA Filter Shock Tube Tests.

Filter Condition	Serial No.	Shock Over Pressure (psig)	Time of Break (msec)
Clean Filter	184697	2.08	Filter did not break
Clean Filter	184733	3.25	9.0

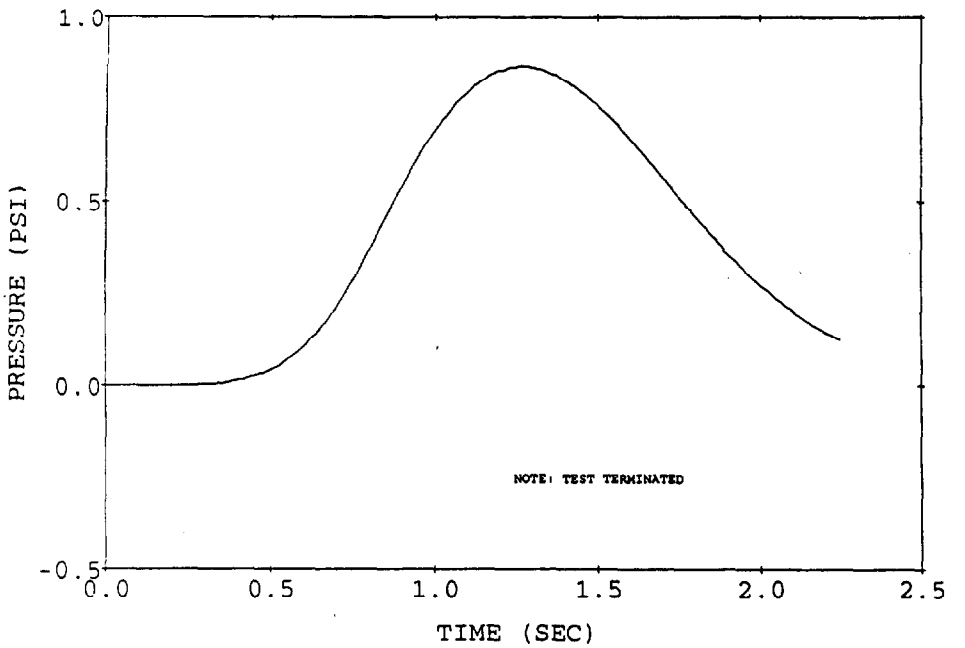

Figure 6 Tornado Pressure Pulse for Test SN185274.

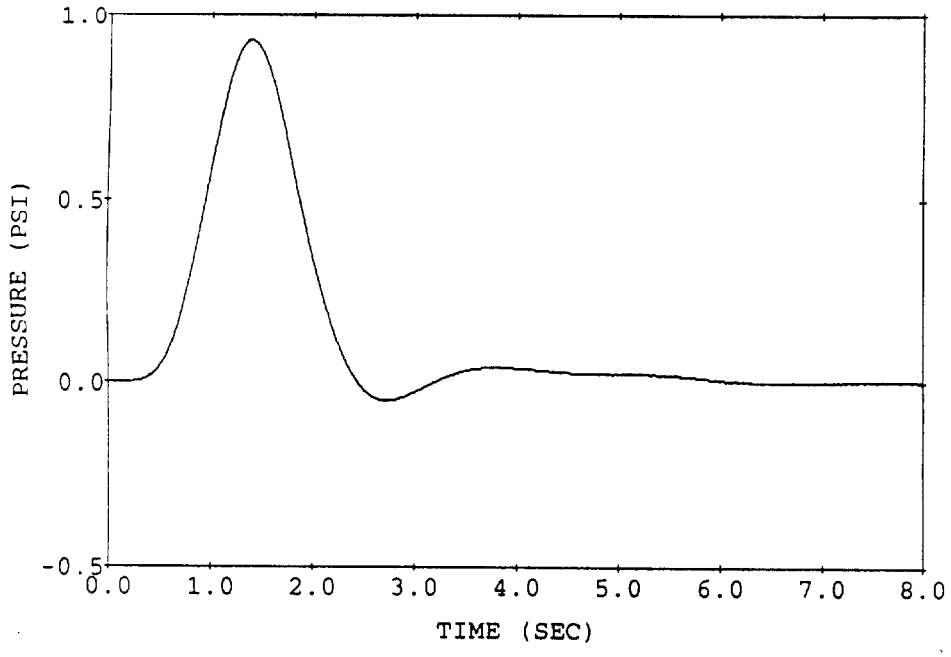

Figure 7 Tornado Pressure Pulse for Test SN184659.

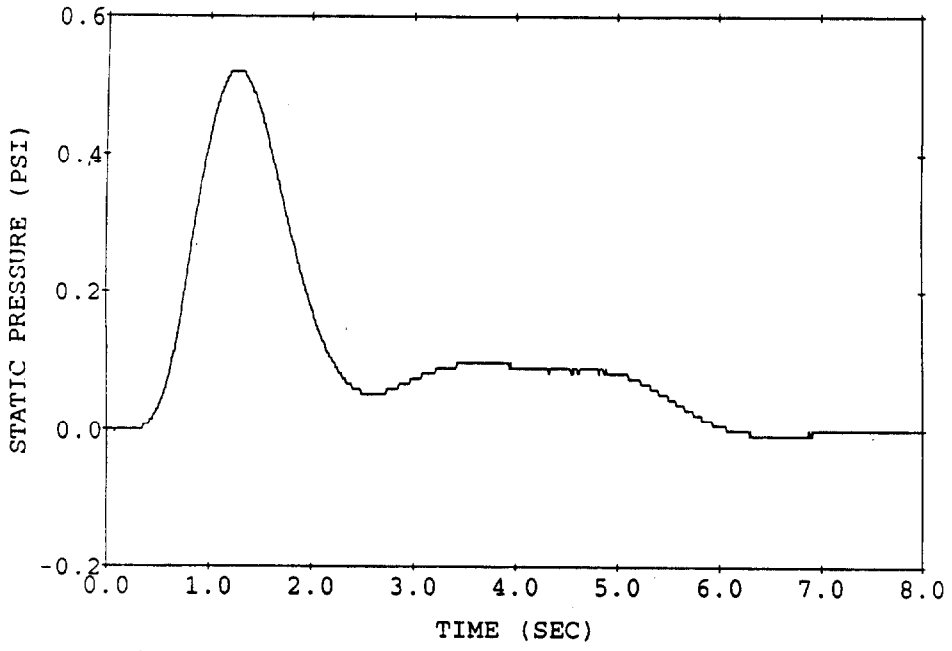

Figure 8 Tornado Pressure Pulse for Test SN184783.

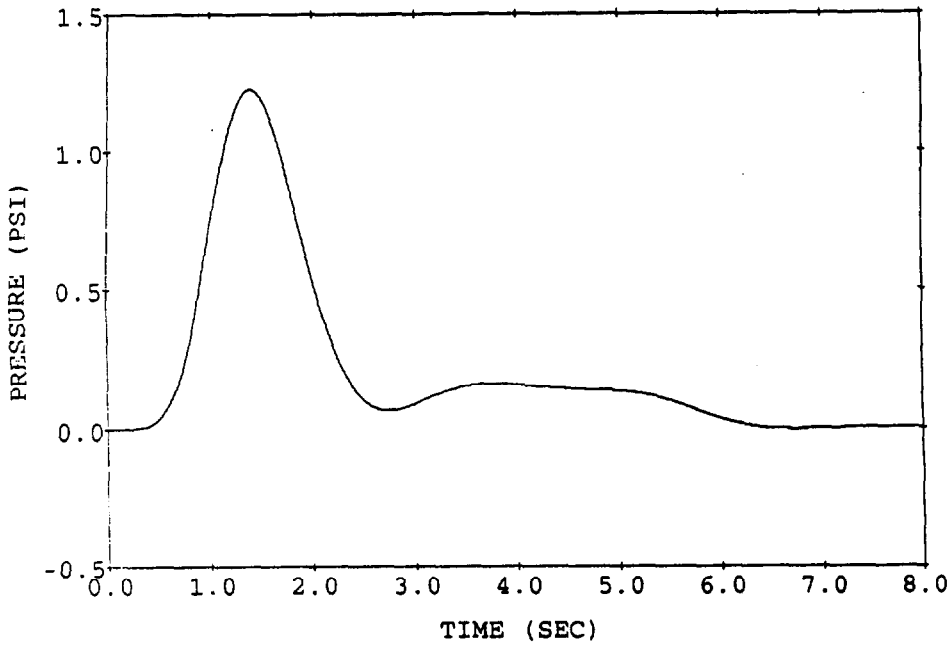

Figure 9 Tornado Pressure Pulse for Test SN184684.

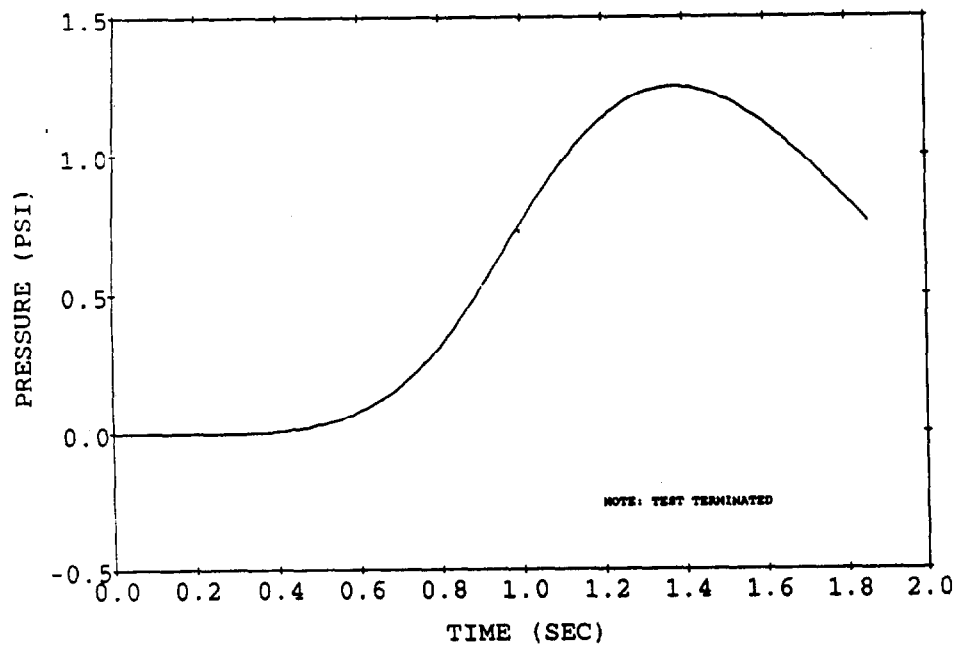

Figure 10 Tornado Pressure Pulse for Test SN185234.

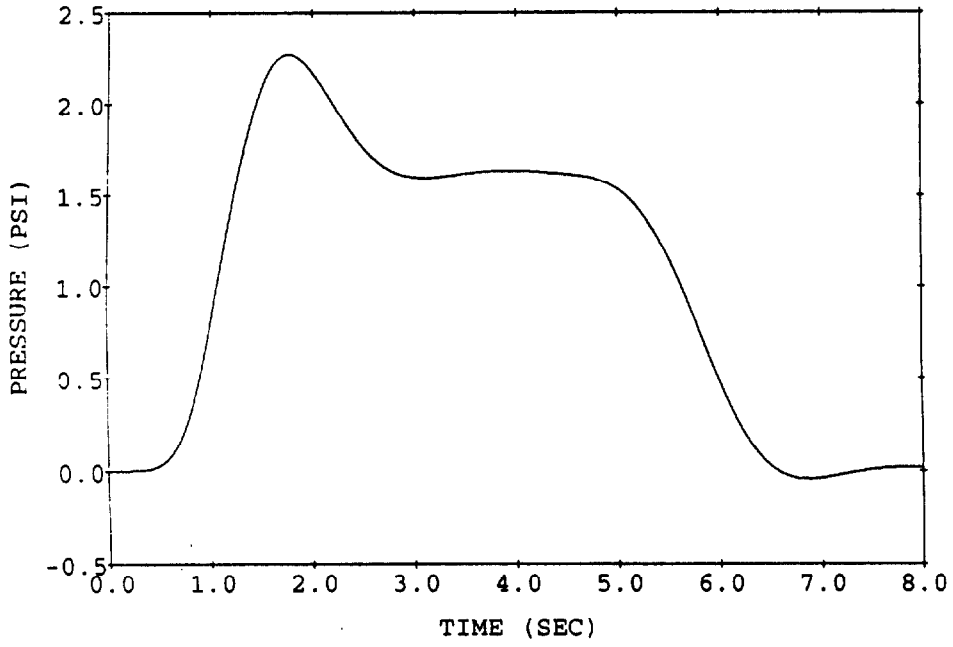

Figure 11 Tornado Pressure Pulse for Test SN184678.

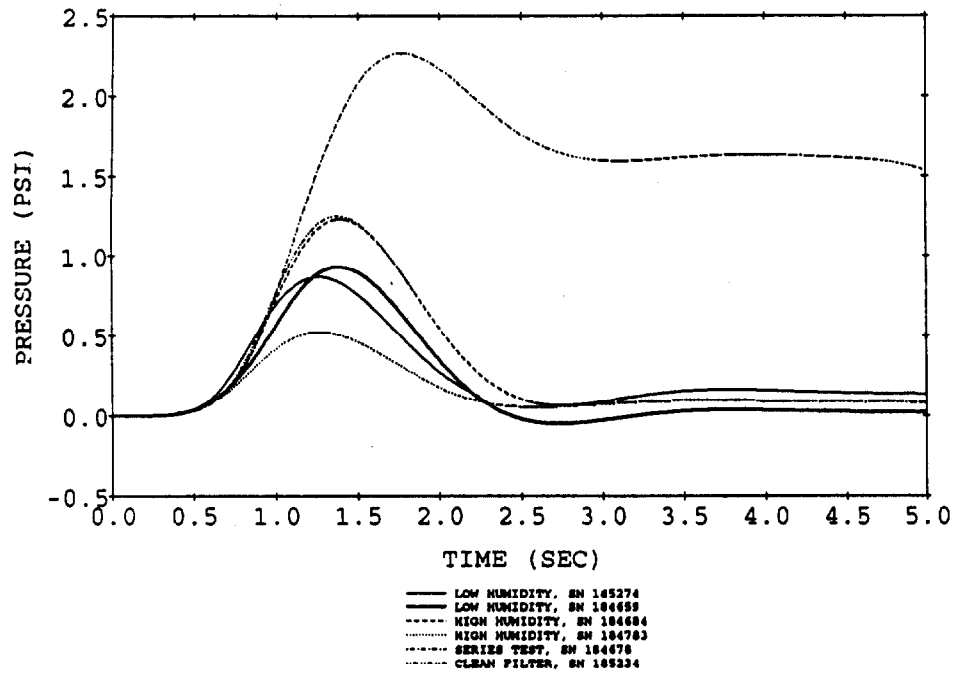

Figure 12 Composite of Figures 6 through 11.

Figure 13 shows the recorded flow rate through a typical filter (SN184684) during the tornado blow down test. The flow rate through this filter reach a maximum of about 50,000cfm or about 50 times its nominal operating flow rate of 1000cfm.

In general it was found that a HEPA filter had a structural strength which depended upon the relative humidity at which it was loaded. If the relative humidity (RH) was maintained at about 13 to 14% during the loading of the salt dust into the filters, then their structural strength was reduced by about 31% from their clean strength. However, when the RH was maintained at about 73%, then the structural strength of the salt loaded filter was essentially that of the clean filters. This can be explained as follows. The filter medium (paper) is the element of the HEPA filter which generally fails first by rupturing or tearing[2,3] when the filter is subjected to a large pressure pulse. So the pressure drop that can be maintained across the filter medium is very important. In the case when a filter is loaded with salt dust at 13 to 14% RH the salt is well below its equilibrium RH of 40%, above which it would liquify. Hence, the salt dust remains dry and no agglomeration of small particles occurs. The very fine dust particles challenging the HEPA filter are thus trapped within the filter medium itself and are responsible for most of the increase in pressure drop across the filter. However, when the RH is above 40%, then the salt dust in the air challenging the filter begins to melt and become liquid droplets. These droplets can agglomerate to cause larger droplets. When the droplets strike the wire screen guarding the upstream face of the HEPA filter they crystalize, forming a nearly continuous crust. Very little of the salt is actually trapped within the filter medium itself. Hence, most of the pressure drop across a HEPA filter loaded with salt dust entrained in air at about 73% RH is due to the crystallized salt upon the upstream wire face guard of the filter. The filter medium, therefore, acts (in its failure) as if it were clean. Figure 14 is a photograph showing the face of a filter loaded with salt under the low humidity condition. Notice that little of the salt has collected on the wire face guard. Figure 15 is a photograph of a filter loaded with salt under the high humidity condition. Notice the large amount of salt crystallized upon the wire face guard.

One structural test was performed on a filter (SN184783) loaded with salt under the high humidity conditions and then pre-conditioned with an air stream of 100% RH for four hours just before structural testing. Water was seen running off the filter medium just before the structural test. The structural strength of this filter proved to be less than 50% of the strength of a clean filter. In this case the liquid water on and within the filter medium apparently weakened the medium.

A structural test was performed on a clean unloaded HEPA filter (SN184678) in series with a pre-filter 1.23m upstream. The pressure drop of the combined filter system was 14.1kPa at failure. The HEPA filter failed completely, but the filter pack of the pre-filter remained in place, as can be seen in the post test photograph shown in Fig. 16. Apparently, the pre-filter protected the HEPA filter since

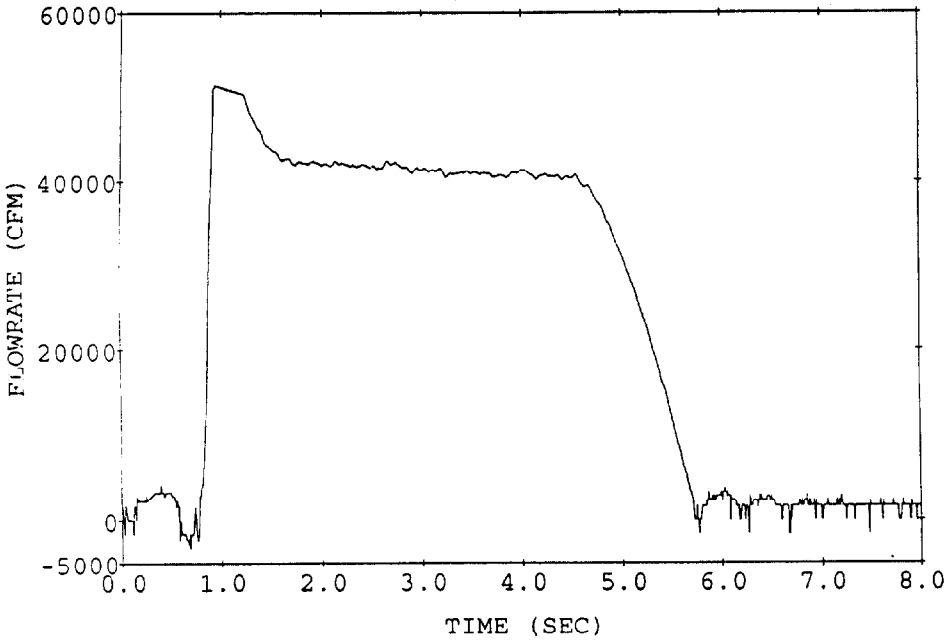

Figure 13 Typical Flow through HEPA Filter During Tornado Pressure Pulse.

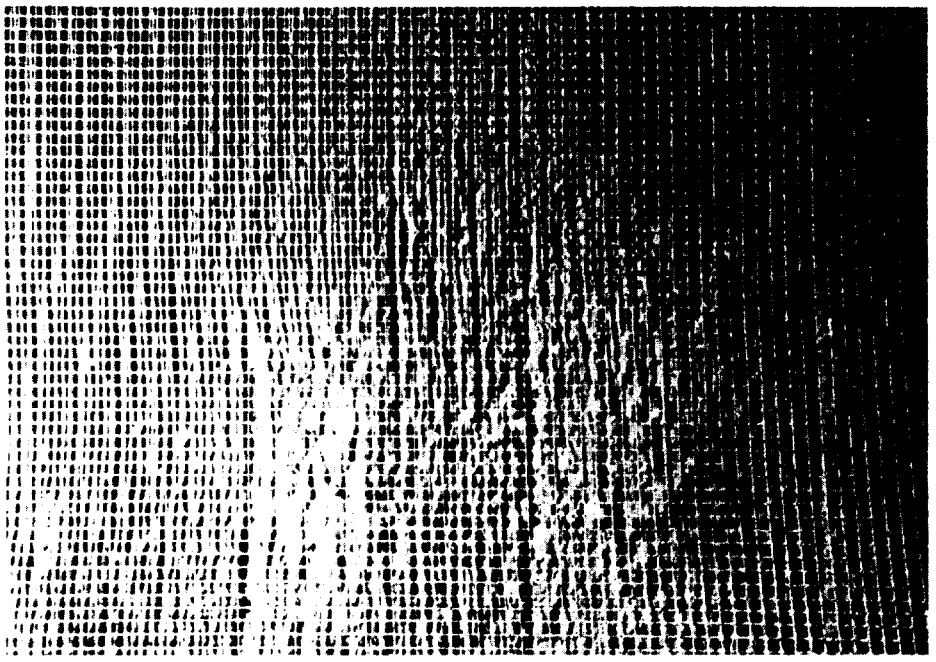

Figure 14 Photograph of Face Guard of HEPA Filter Loaded Under Low Humidity Conditions.

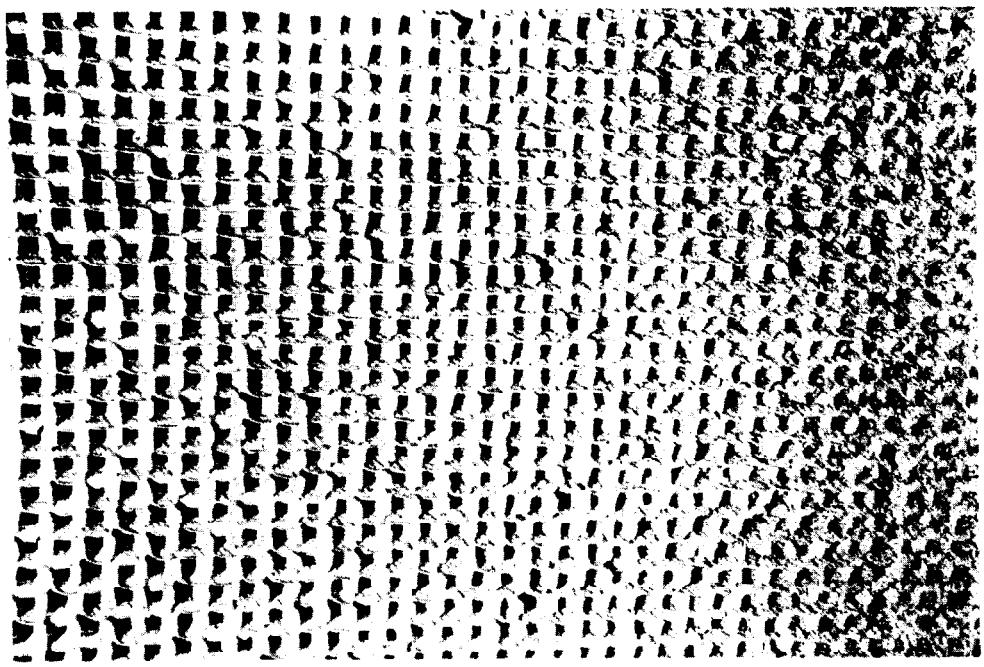

Figure 15 Photograph of Face Guard of HEPA Filter Loaded Under High Humidity Conditions.

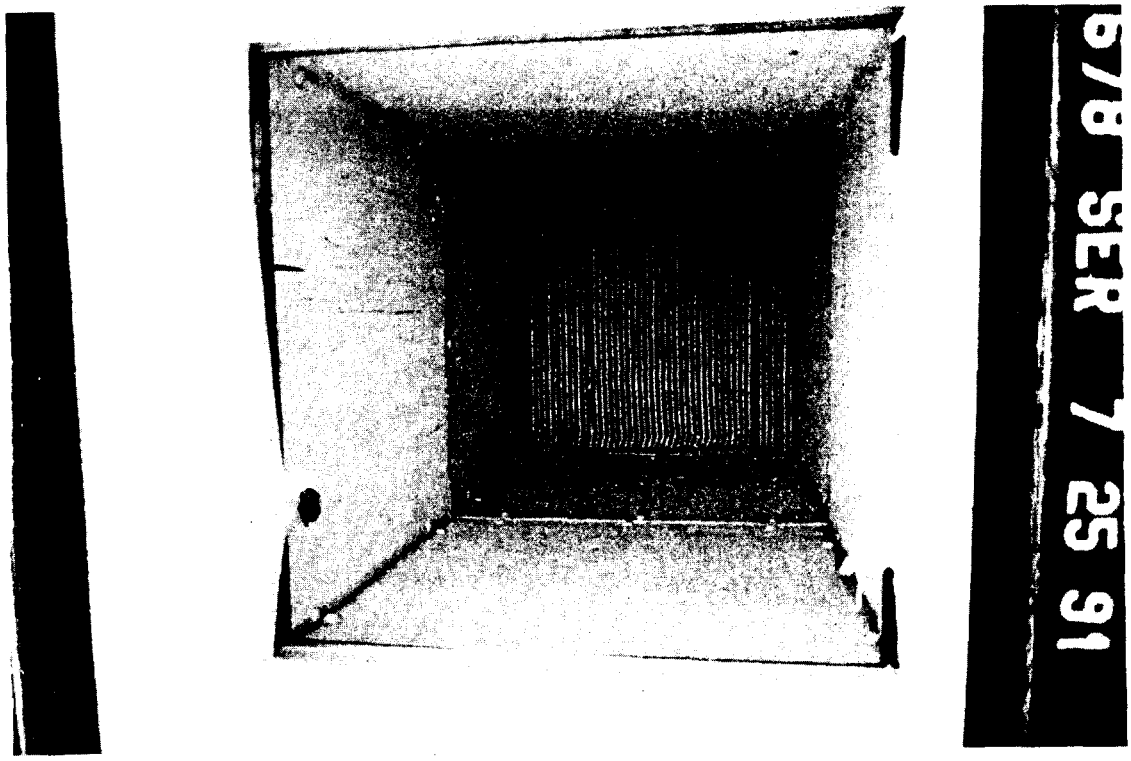

Figure 16 Post Test Photograph of HEPA Filter and Pre-Filter in Series. Filter Pack of the HEPA Filter (Foreground) has been Blown Out.

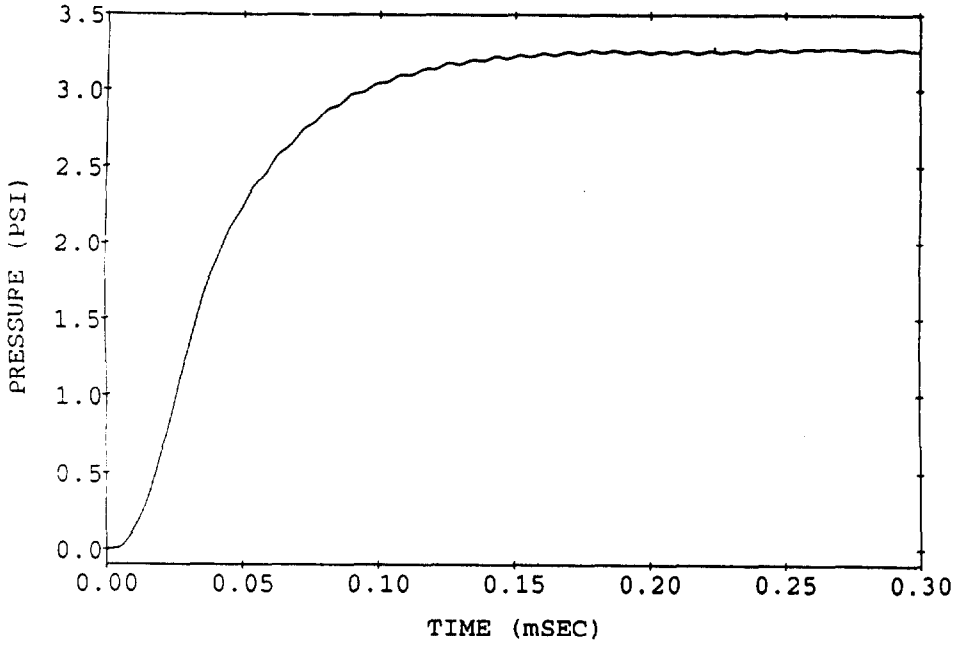

Figure 17 Shock Pulse for Test SN194733.

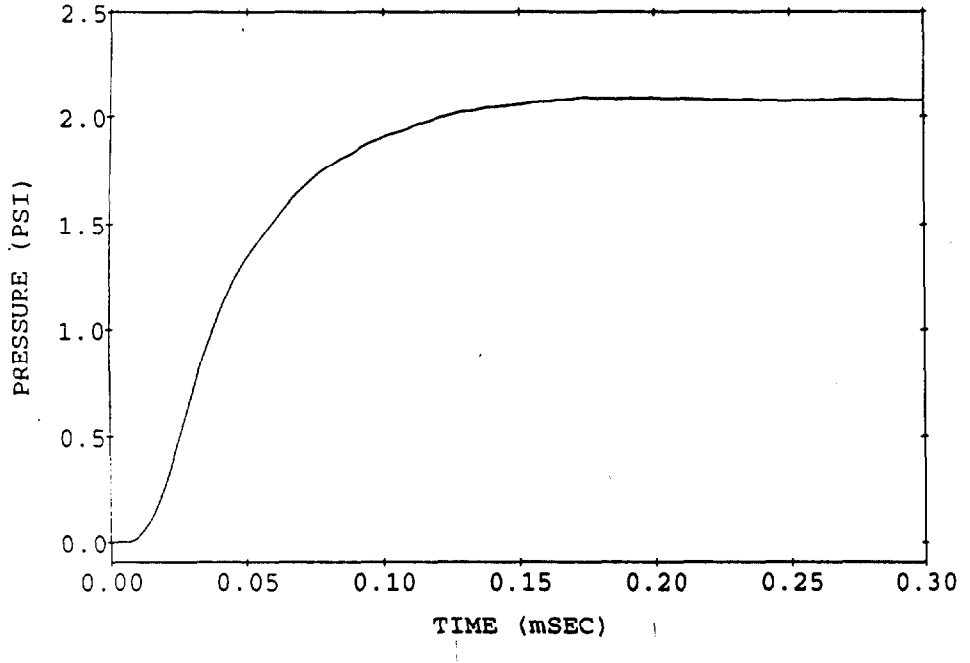

Figure 18 Shock Pulse for Test SN184697.

the total pressure drop at failure was nearly twice that of the pressure drop at failure for a single HEPA filter (SN185234).

The pressure drop that caused failure of HEPA filters was about 8.24kPa for a clean filter and for a filter loaded with salt dust at 73% RH. This agrees well with the results of Ricketts[11] who tested a large number of the same brand of HEPA filters. Note that this failure pressure is well below the maximum 3psi pressure drop that might be expected from an NRC Region I tornado. Hence, there is a strong possibility that these HEPA filters would fail structurally if a tornado were to pass over the WIPP ventilation building.

Shock Overpressure Tests: Table 2 summarized the results of the HEPA filter shock tests which were performed. Two clean, unloaded 0.6m by 0.6m by 0.3m HEPA filters from the WIPP site were tested. Figures 17 and 18 show the shock overpressures experienced by each filter. The overpressure of 22.3kPa for filter SN184733 caused it to just break at 9msec after the shock wave struck the upstream face of the filter. It has been shown by Gregory, Martin, Smith, and Fenton[12] that the impulse of a shock wave is what causes a HEPA filter to fail. The calculated impulse per unit area in our case is

$$\frac{I}{A}=\int p\,dt=3.25X9=29.25\,psi\text{-}msec=201\,kPa\text{-}msec$$

This value was within 10% of the value given in ref. 12 for the same brand of HEPA filter tested for this study.

IV. Conclusions

The following conclusions can be drawn:

1. The structural strength of HEPA filters loaded with salt under low humidity conditions (13-14%RH) was reduced by about 31% from their clean strength, when the filters were subjected to a tornado-like pressure pulse.

2. The structural strength of HEPA filters loaded with salt under high humidity conditions (73%RH) was not significantly reduced from their clean strength when subjected to a tornado like pressure pulse.

3. Salt crystallized on the wire face guards of the HEPA filters loaded with salt under high humidity conditions and caused the majority of the pressure drop across the filters.

4. The filter medium of the HEPA filters loaded with salt under high humidity conditions did not collect as much salt particulate as

did the filter medium of the HEPA filters loaded under the low humidity conditions.

5. The structural strength of the HEPA filters conditioned at 100%RH for 4 hours before structural testing was less than 50% of clean, dry strength, implying that filter medium saturated with water looses much of its strength.

6. A clean HEPA filter in series with a pre-filter can withstand almost twice the magnitude of a tornado-like pressure pulse than can a single clean HEPA filter, but it will still fail at pressure differences less than those of the Region I NRC tornado pulse.

7. A clean HEPA filter failed at a shock impulse of 201kPa-msec.

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CLOSING COMMENTS OF SESSION CO-CHAIRMAN MOELLER

This session has been interesting and stimulating. The first two papers in Session 5 covered new developments on the measurement and removal of radionuclides in airborne effluent streams generated as a result of the vitrification of high level radioactive wastes. In the initial paper, scientists from Japan reported on a perforated plate column scrubber for removing ruthenium from off-gas streams. In the second paper, workers at Savannah River reported on the development of a new technique, using inductively coupled plasma-mass spectrometry, to measure non-radioactive cesium in their off-gas system and thus provide a method that is sufficiently sensitive to assess the decontamination factors for cesium in the melter. The third and last paper in the session summarized structural tests developed for the evaluation of HEPA filters to be used in the emergency ventilation system at WIPP. Interestingly, these tests showed that filters loaded under high humidity had greater structural strength; presumably this is due to the crystallization of salt upon the wire face guard of the filter.